Outstanding Issues in Polar Ozone Loss

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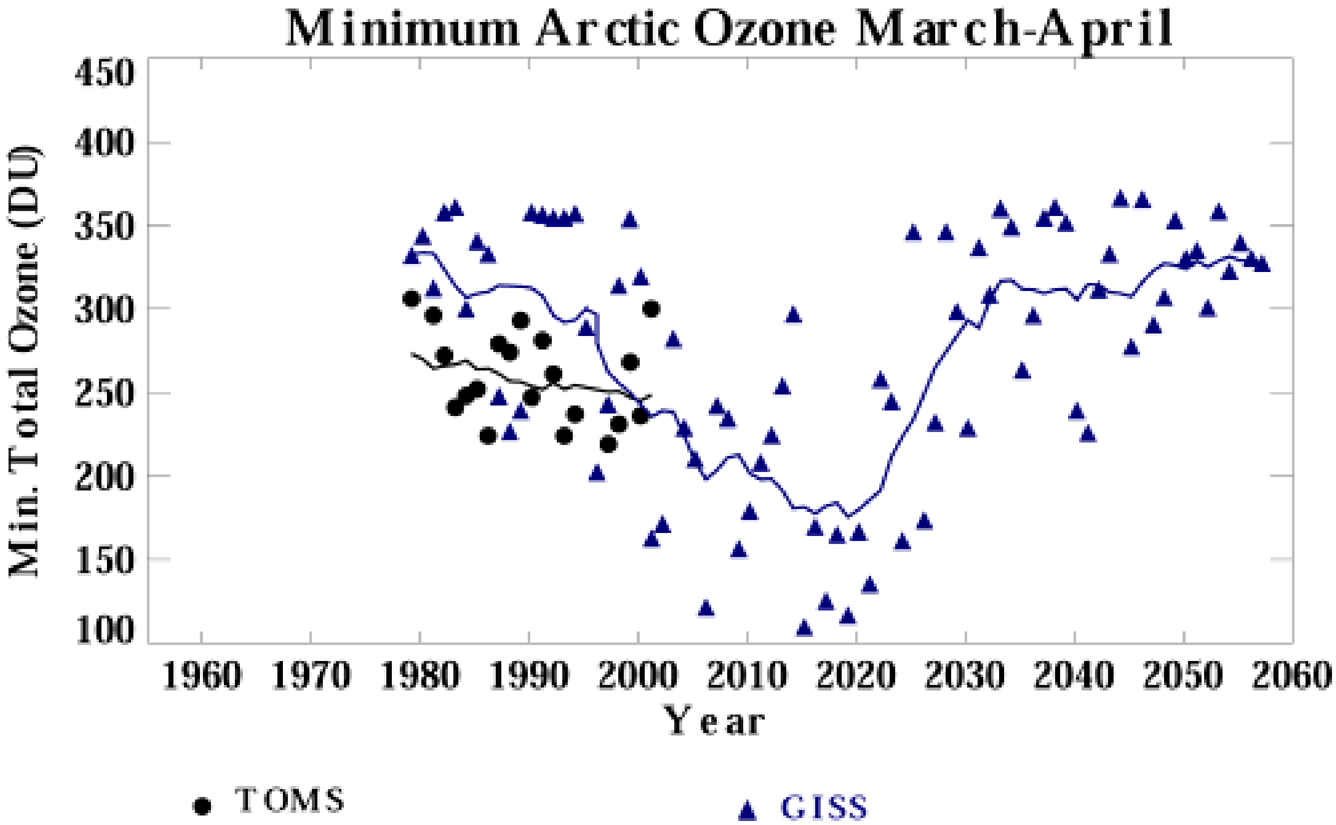
SOSST Meeting, June 2004

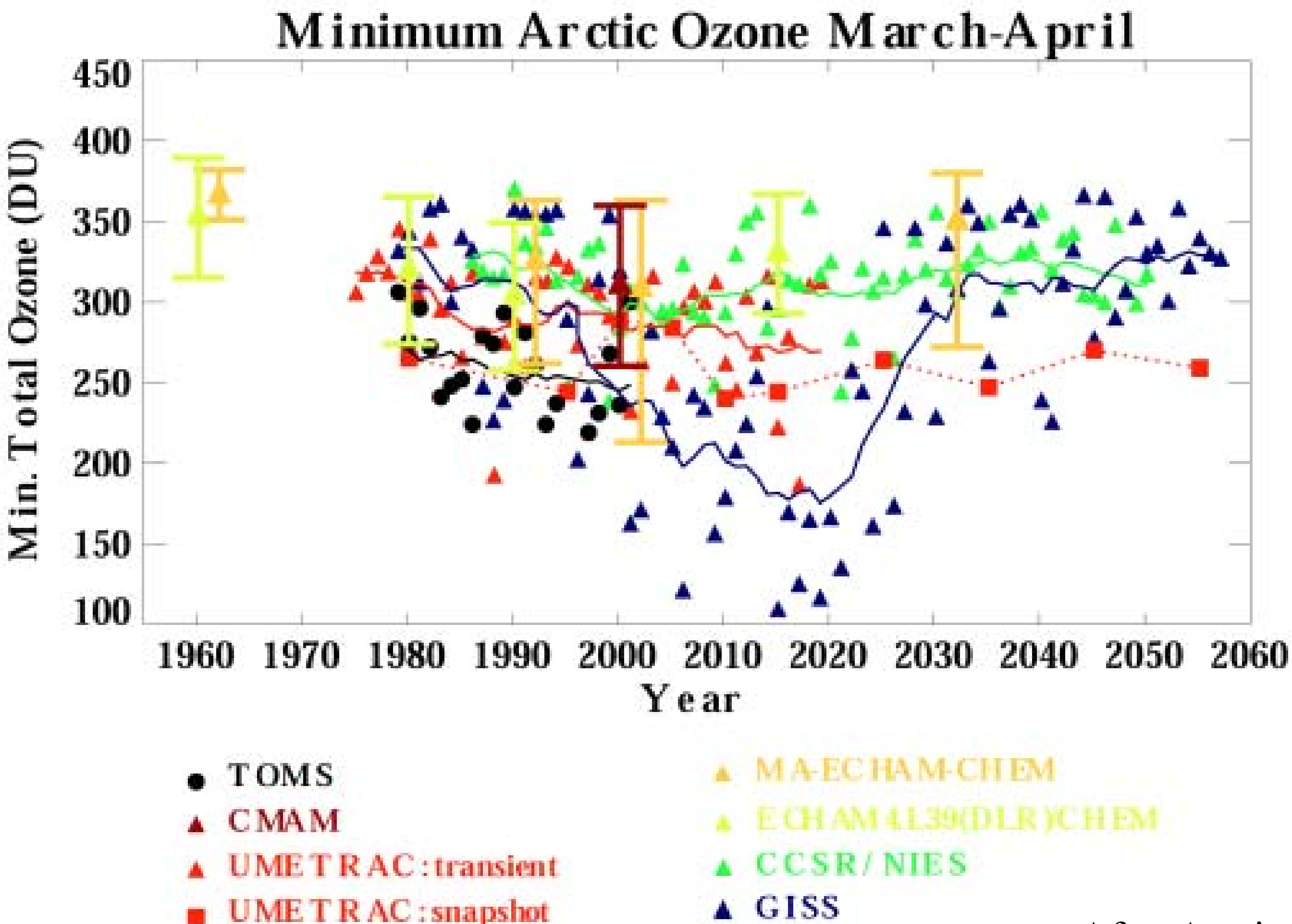
Future Evolution of Arctic Ozone – GISS Model



After Shindell et al., Nature, 1998

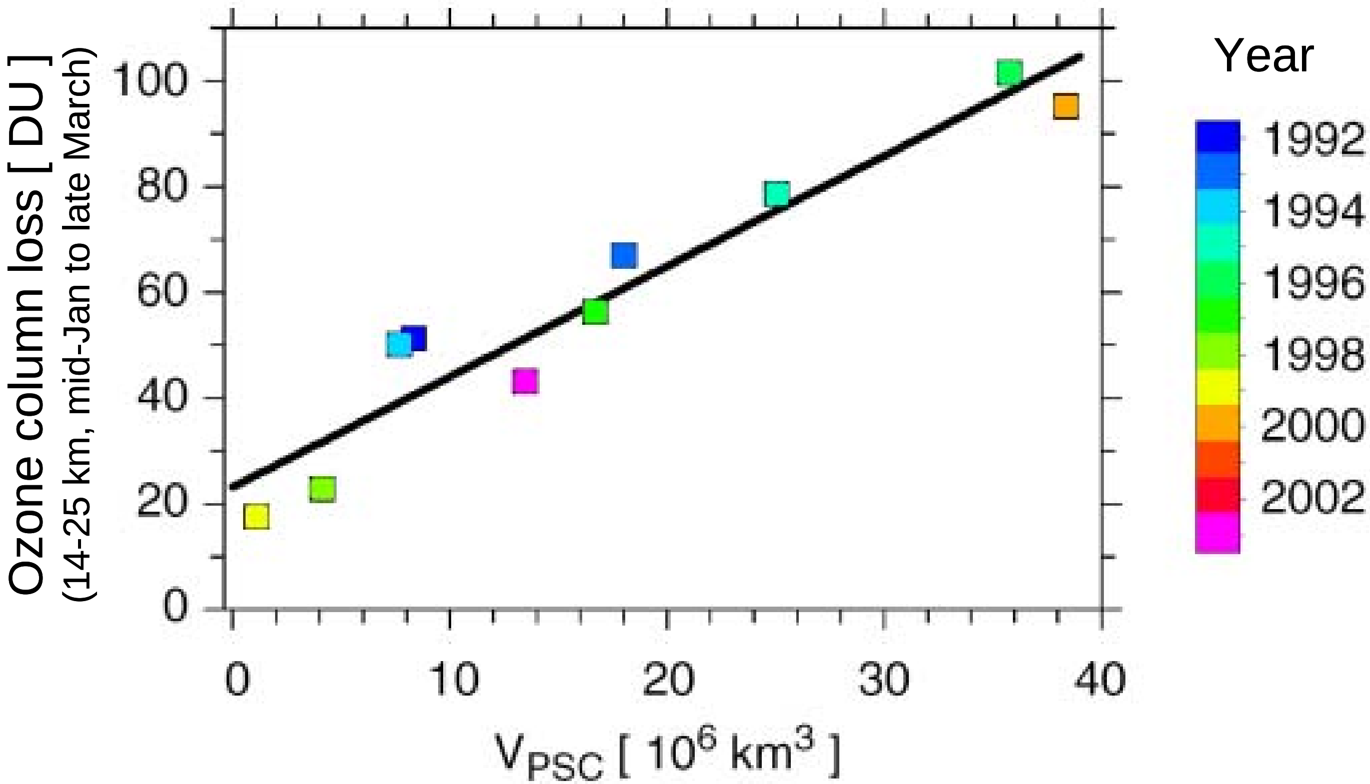
Future Evolution of Arctic Ozone – Many Models

CCMs – Chemistry-Climate Models



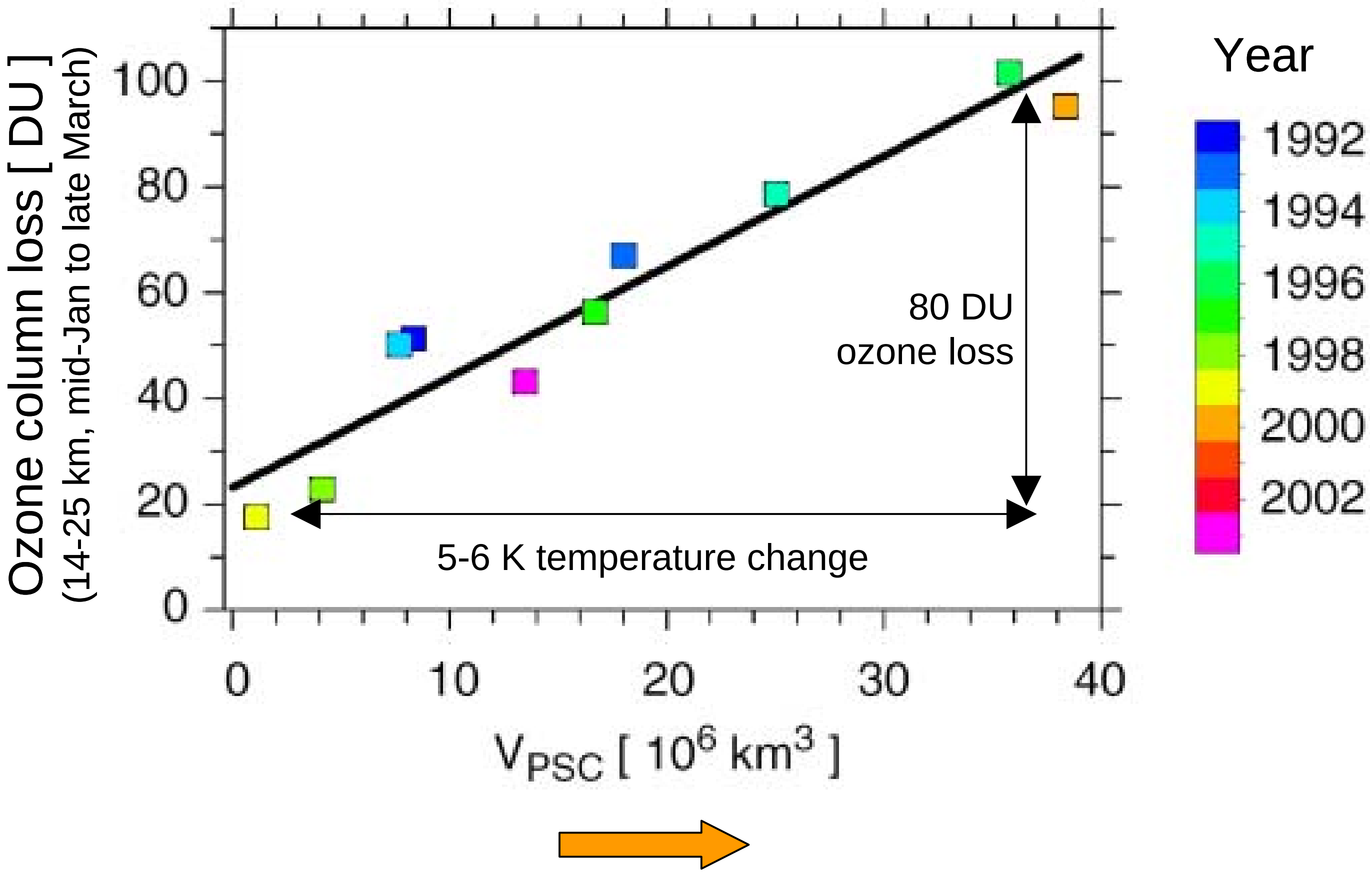
After Austin et al., ACP, 2003

Ozone Loss Versus V_{PSC}



Rex et al., GRL, 2004

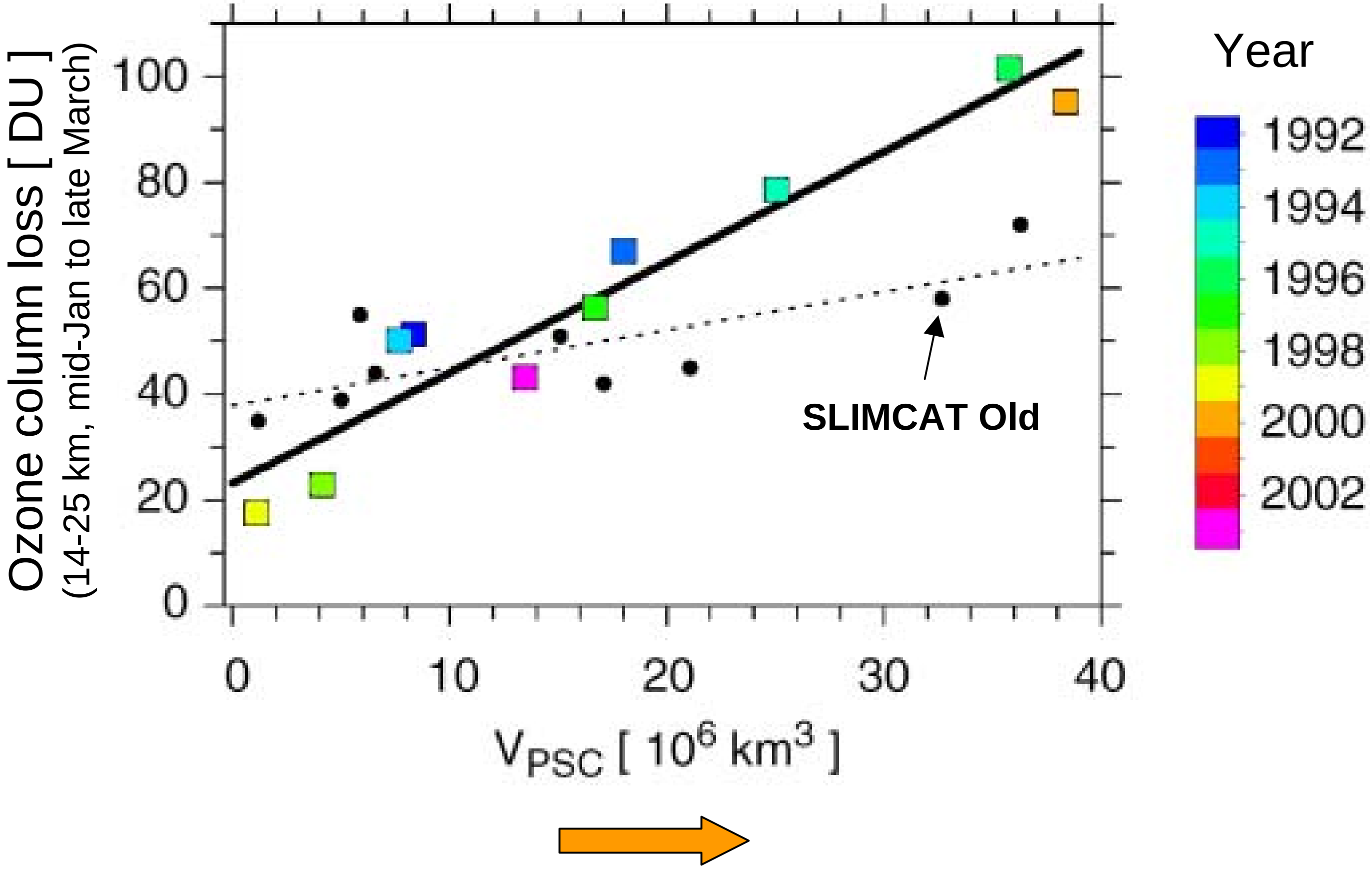
Impact of Climate Change on Arctic Ozone Loss



~ 15 DU additional ozone loss
per Kelvin cooling of the Arctic stratosphere

Rex et al., GRL, 2004

Comparison with SLIMCAT – Old Version



SLIMCAT "Old" underestimates sensitivity of Arctic ozone loss to climate change by a factor of three

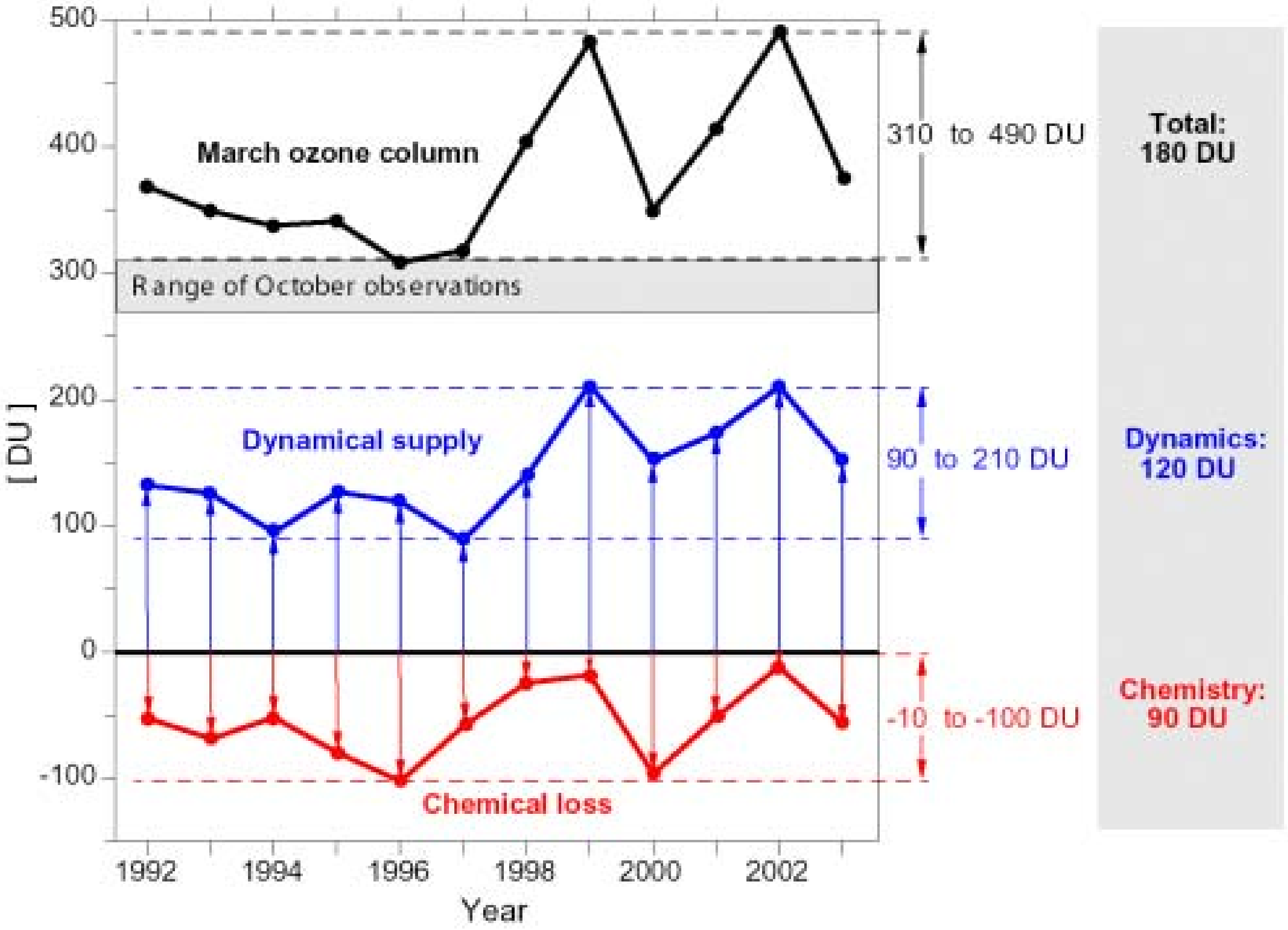
Rex et al., GRL, 2004

Relative Influence of Chemistry and Transport on Arctic Ozone Trends: Model

"Using a state-of-the-art three-dimensional stratospheric chemistry-transport model [e.g., "SLIMCAT Old"], we find that north of 63ºN, on average, dynamical variations dominate the inter-annual variability of total column ozone, with little evidence for a trend towards more wintertime chemical depletion of ozone"

Chipperfield and Jones, Nature, 1999

Relative Influence of Chemistry and Transport on Arctic Ozone Trends: Data



What's Wrong with "SLIMCAT Old" ?

1) Underestimates rate of chemical ozone loss:
   - ClO – ClOOCl kinetics
   - BrO abundance

2) Underestimates denitrification

3) Are problems with "SLIMCAT Old" typical of all CCM models ?
   - Need to look "inside" CCMs
   - First Step: CCM Validation Meeting, Garmisch, Nov 2003
     Chipperfield and Salawitch "leads" for chemistry validation

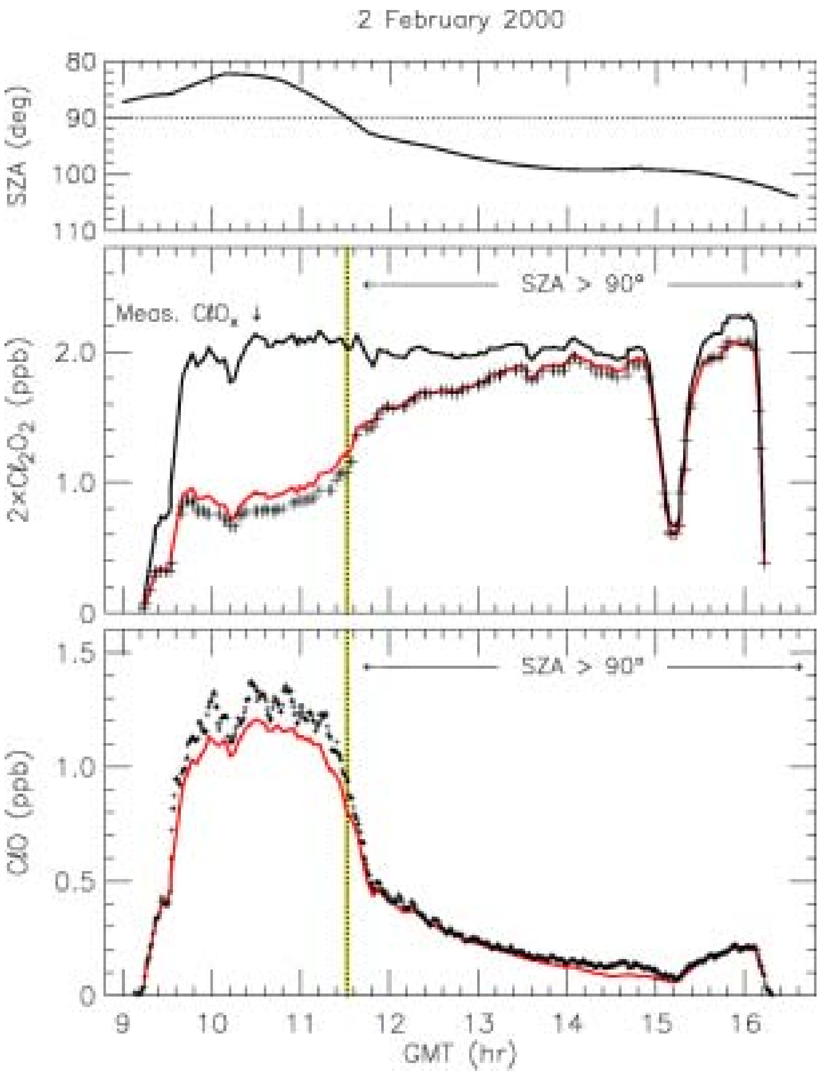
Measured and Modeled ClO-ClOOCl : JPL 2000 Kinetics

$$k_F\,[\mathrm{ClO}]\,[\mathrm{ClO}] \approx J\,[\mathrm{ClOOCl}]$$

$$\Rightarrow\ [\mathrm{ClO}]\,[\mathrm{ClO}] / [\mathrm{ClOOCl}] \approx J / k_F$$

$$\beta\ \text{Ratio} = \frac{[\,\mathrm{ClO}\ \textit{model} \times \mathrm{ClO}\ \textit{model}\,] / \mathrm{ClOOCl}\ \textit{model}}{[\,\mathrm{ClO}\ \textit{meas} \times \mathrm{ClO}\ \textit{meas}\,] / \mathrm{ClOOCl}\ \textit{meas}}$$

$$\approx \frac{(J / k_f)\ \textit{model}}{(J / k_f)\ \textit{actual}}$$



Stimpfle et al.. JGR, 2004

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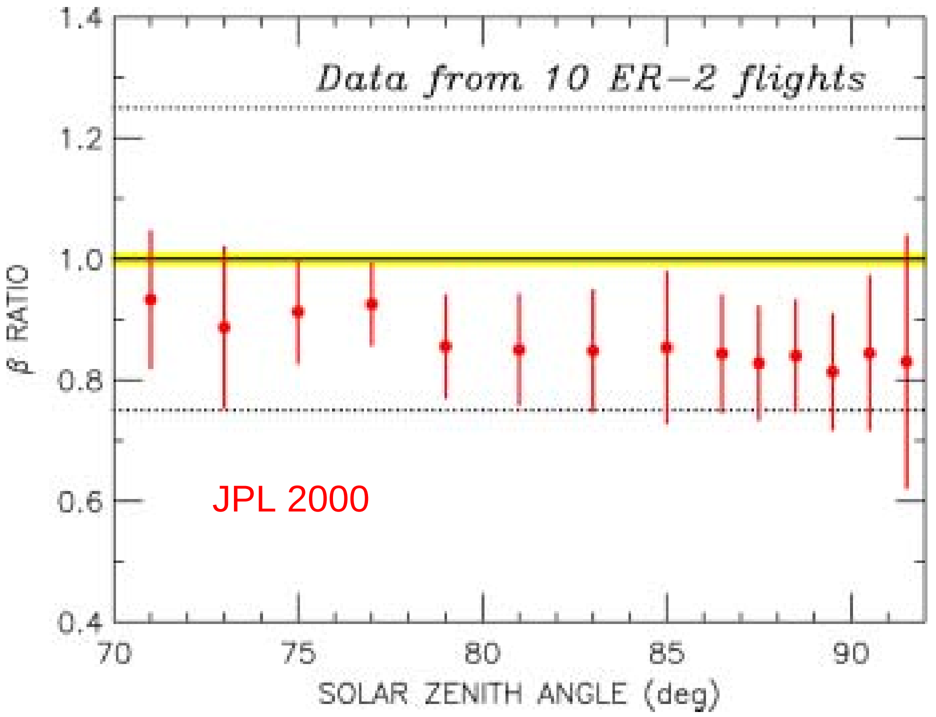

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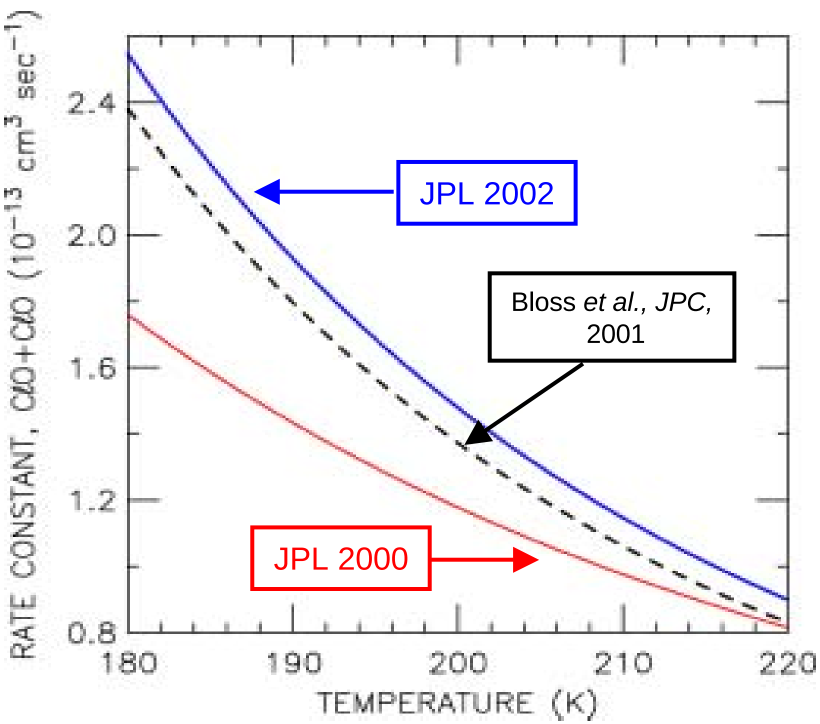

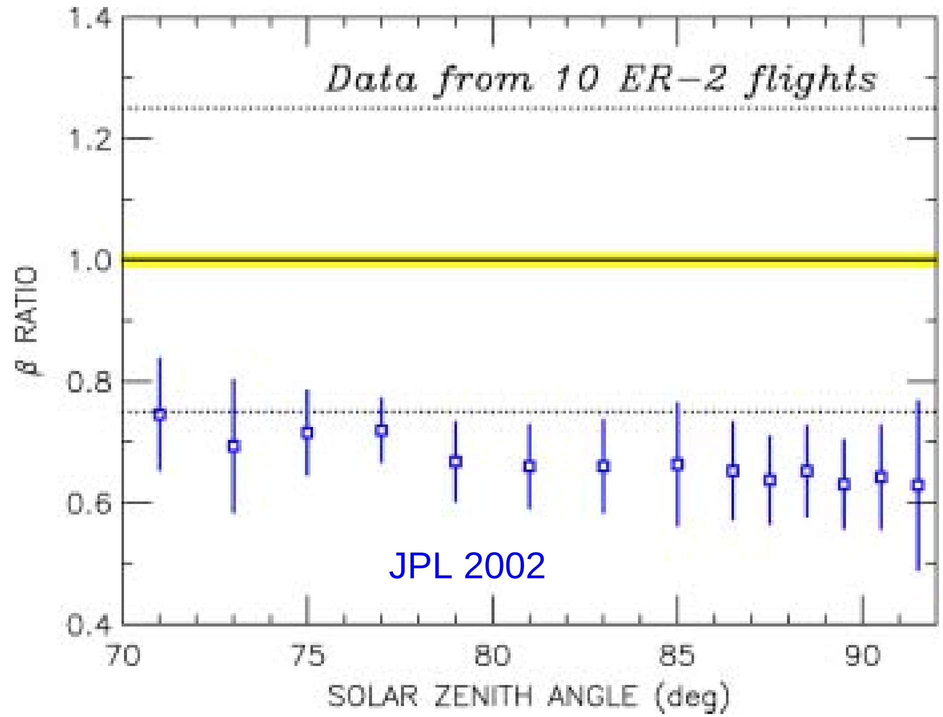

Stimpfle et al.. JGR, 2004

Measured and Modeled ClO-ClOOCl : JPL 2002 Kinetics + Burkholder Cross Section

$$\beta \text{ Ratio} = \frac{[\, \text{ClO}\ \textit{model} \times \text{ClO}\ \textit{model}\,] \,/\, \text{ClOOCl}\ \textit{model}}{[\, \text{ClO}\ \textit{meas} \times \text{ClO}\ \textit{meas}\,] \,/\, \text{ClOOCl}\ \textit{meas}}$$

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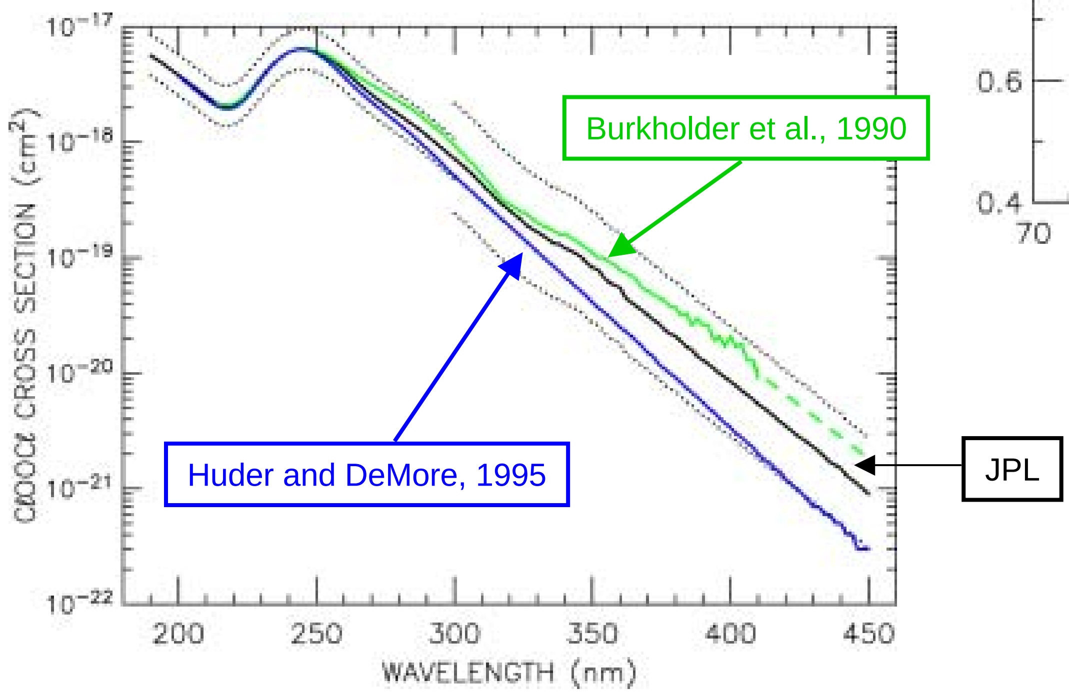
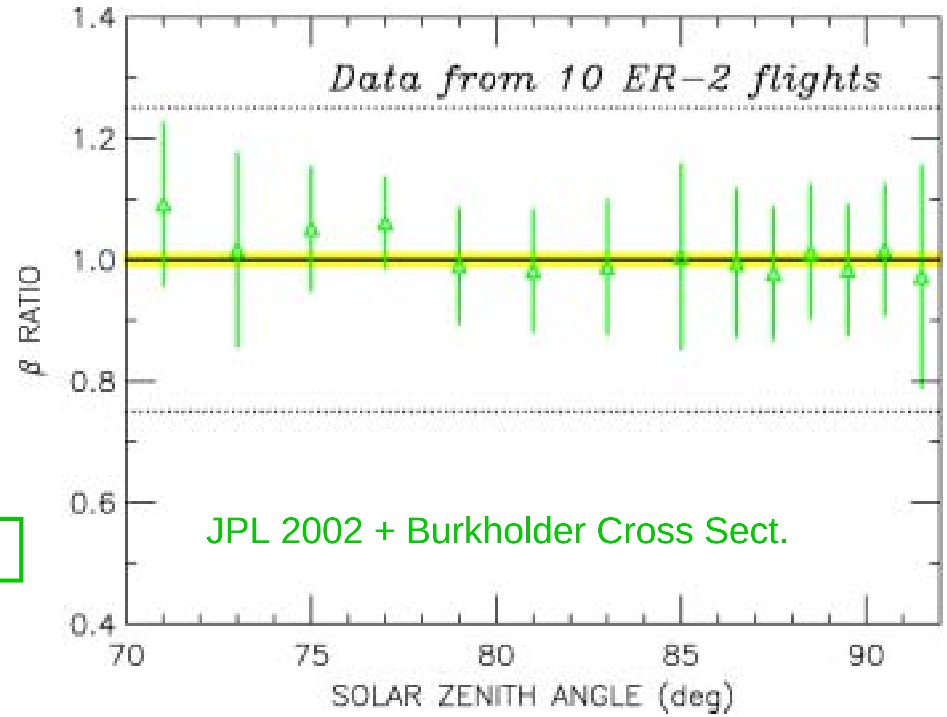
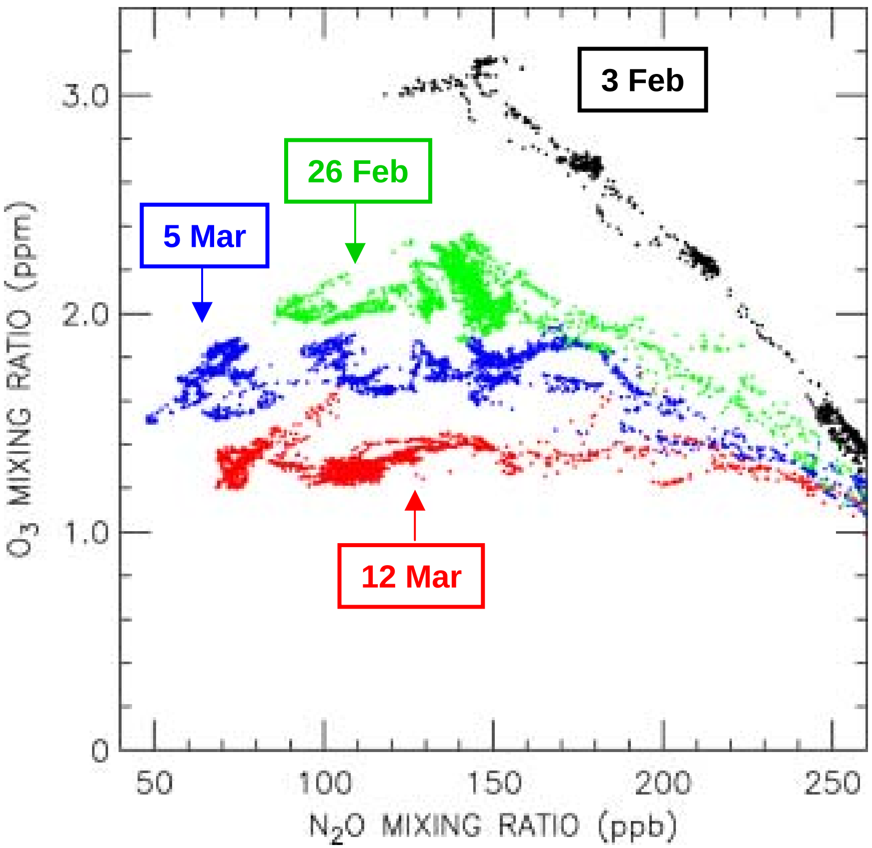


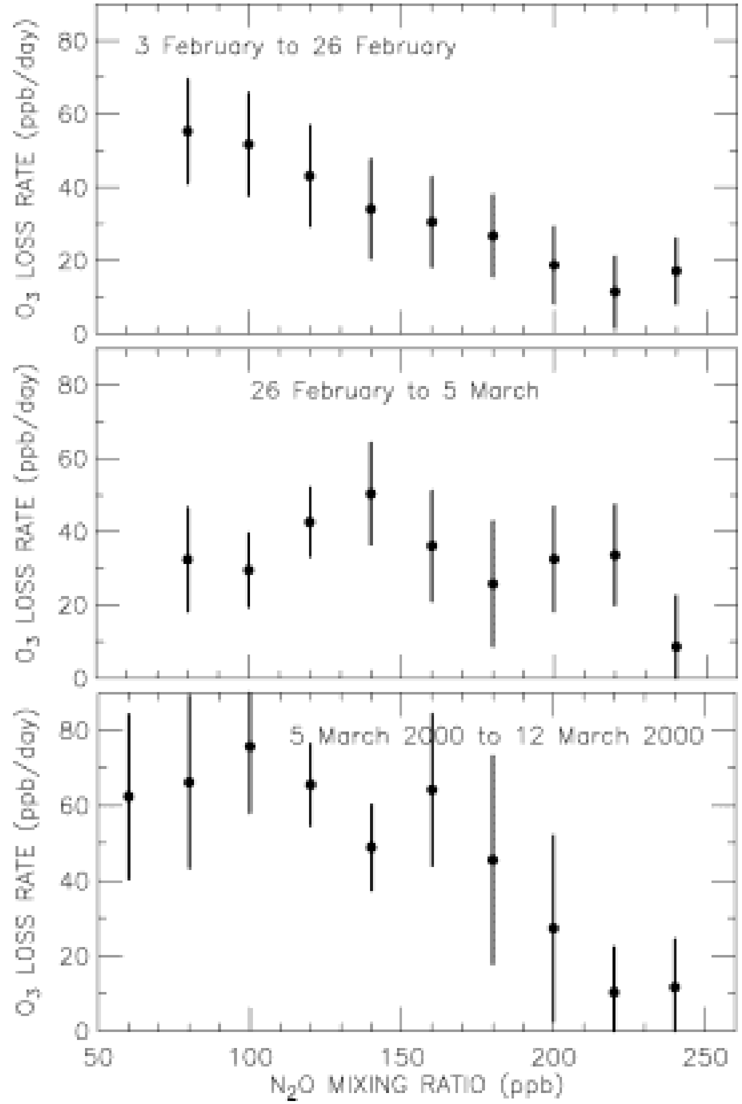


Stimpfle et al.. JGR, 2004

Chemical Ozone Loss Rates: Measured

Evolution of O_3 vs N_2O (*below*) used to define measured ozone loss for three time periods (*right*)





See Richard *et al.*, *GRL*, 2000, Hoppel *et al.*, *JGR*, 2002, Rex *et al.*, *JGR*, 2002 & Salawitch *et al.*, *JGR*, 2002 for demonstrations of the validity of this approach for accurately quantifying observed chemical ozone loss rates.

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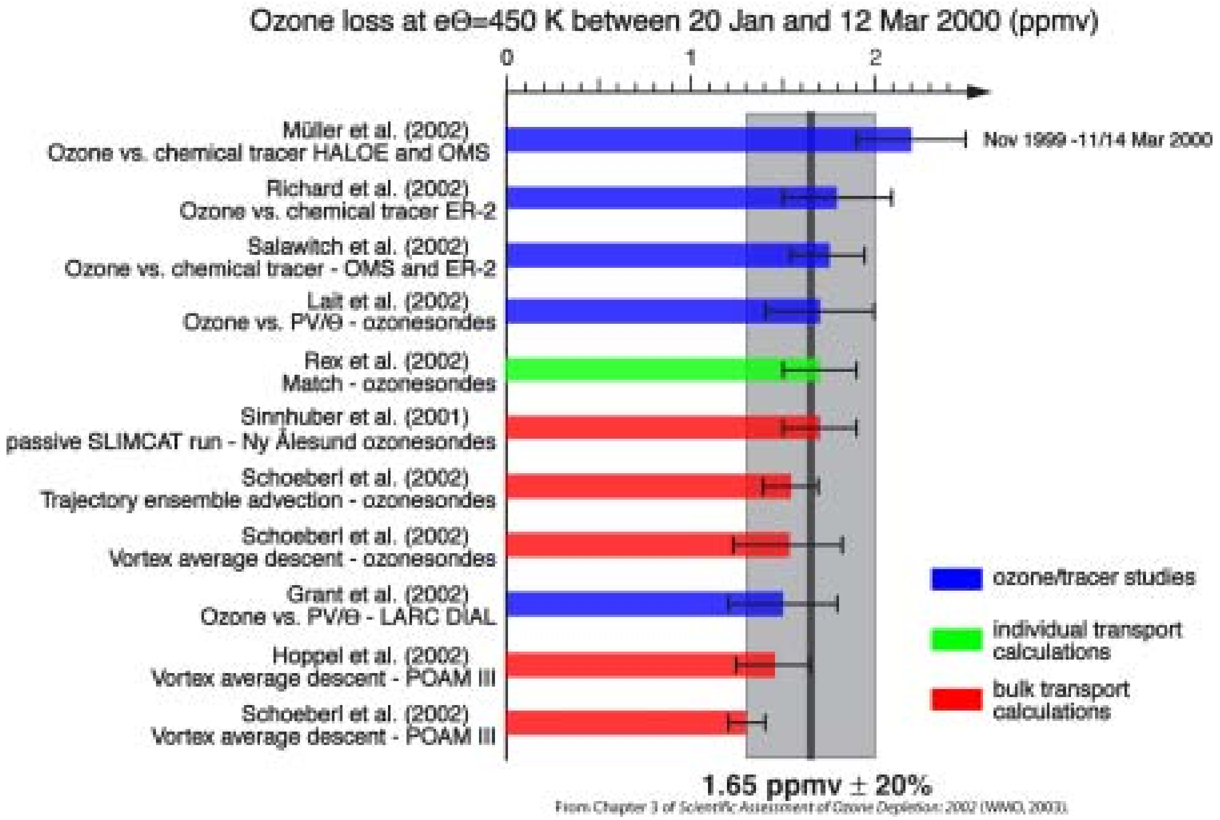
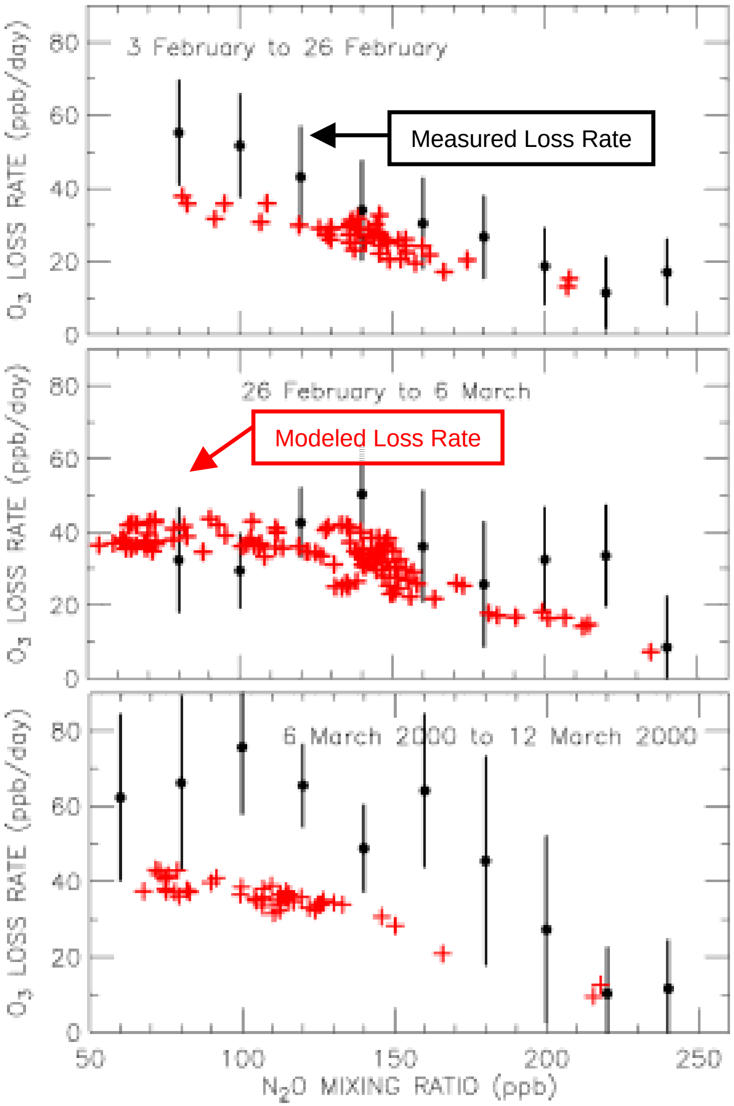
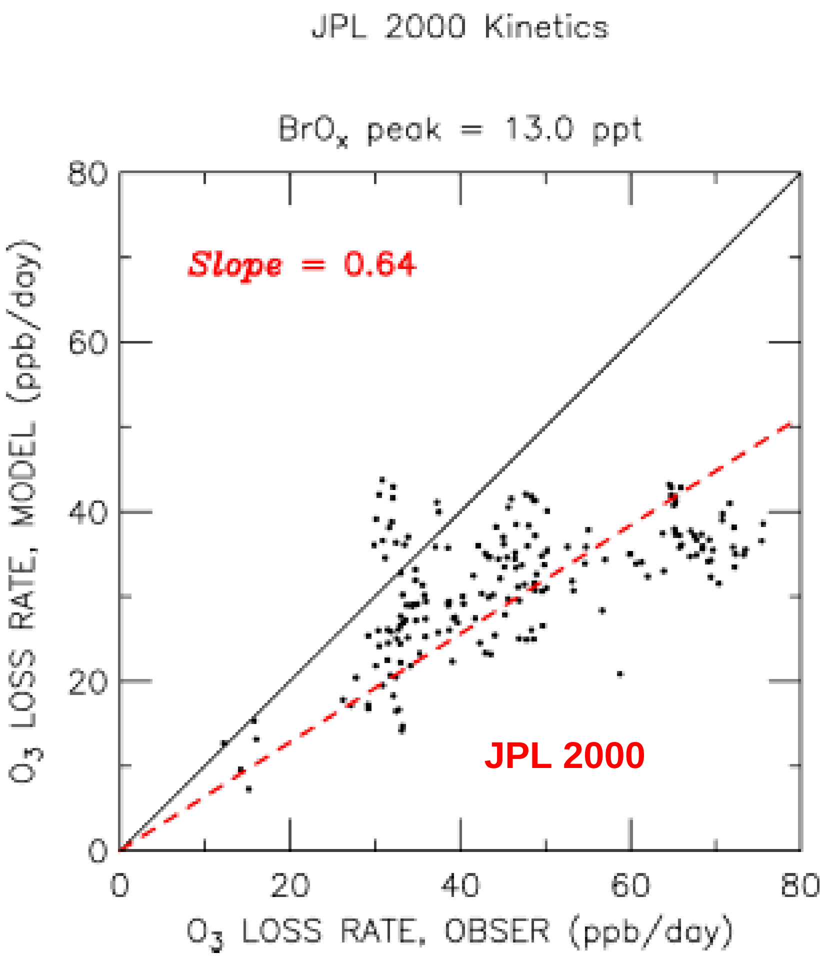


Figure 3-26, WMO 2003

See also Richard *et al., GRL*, 2000, Hoppel *et al., JGR*, 2002, Rex *et al., JGR*, 2002 & Salawitch *et al., JGR*, 2002 for demonstrations of the validity of various approaches for accurately quantifying observed chemical ozone loss rates.

Measured and Modeled Ozone Loss Rates





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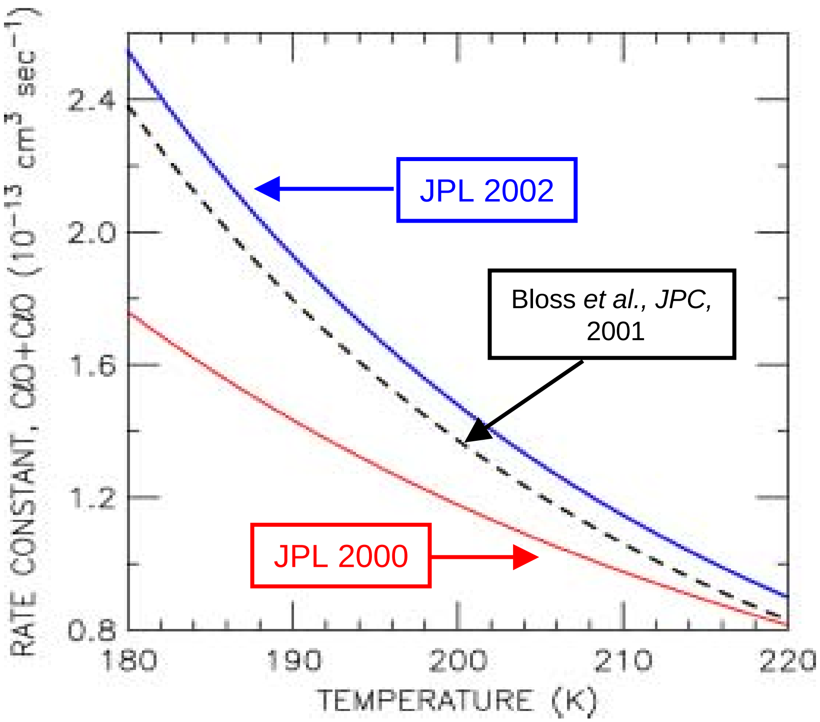
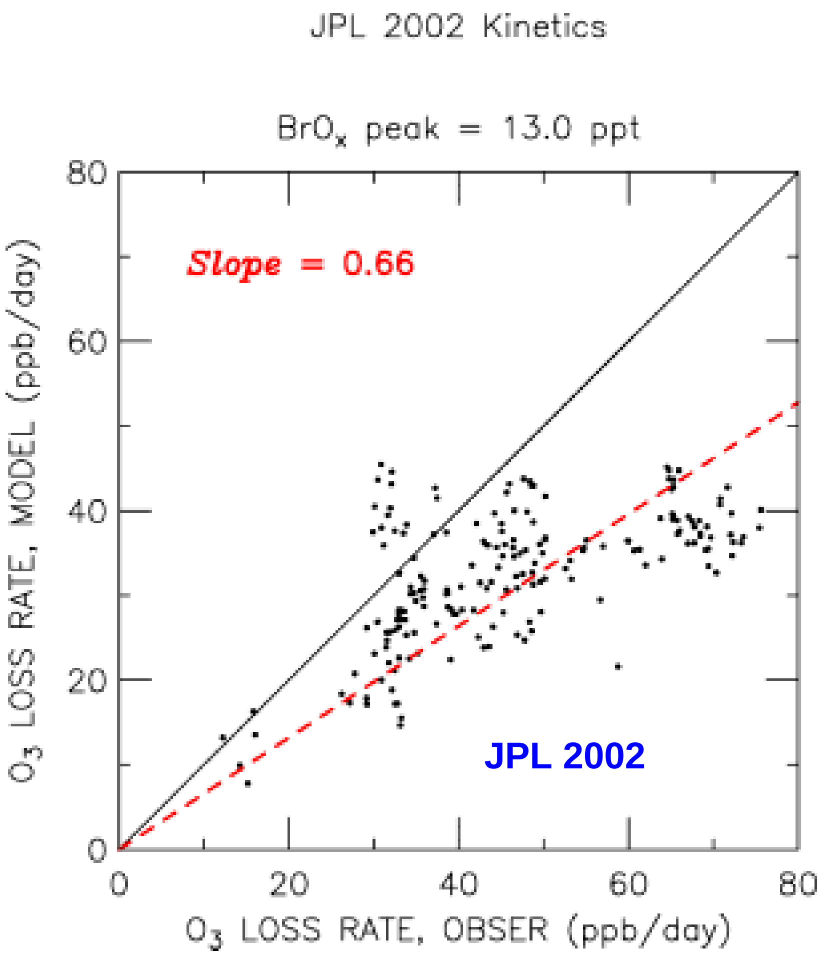
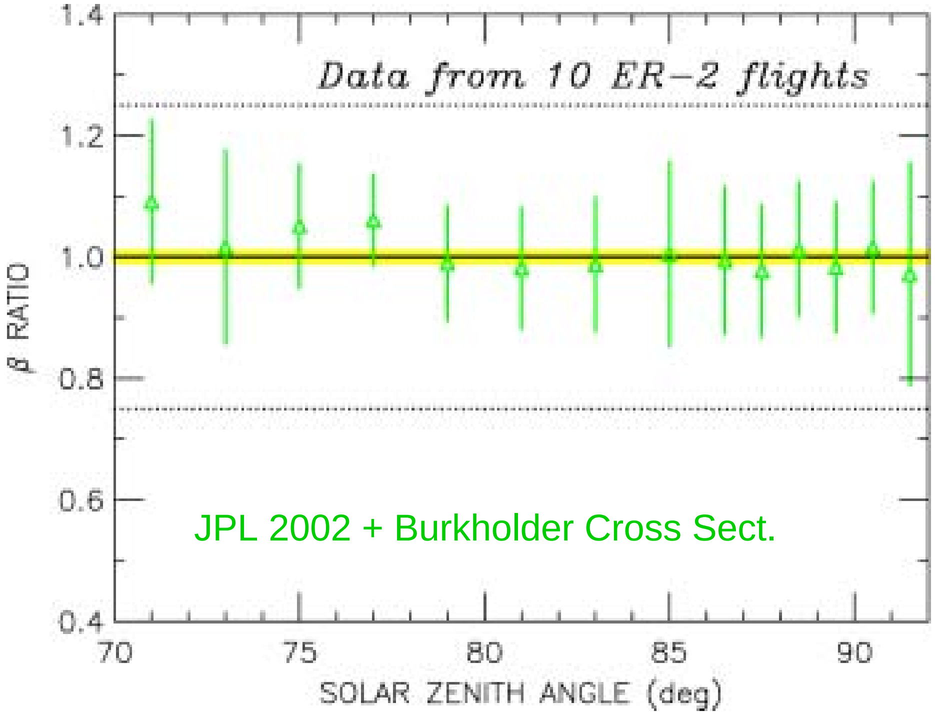
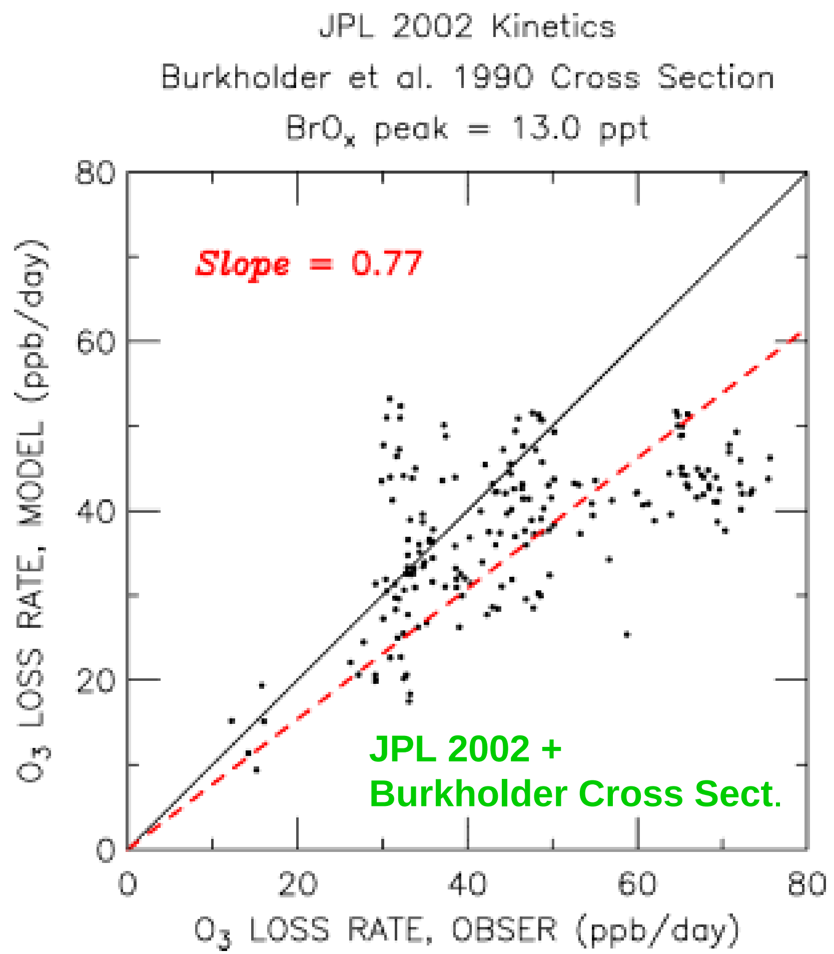




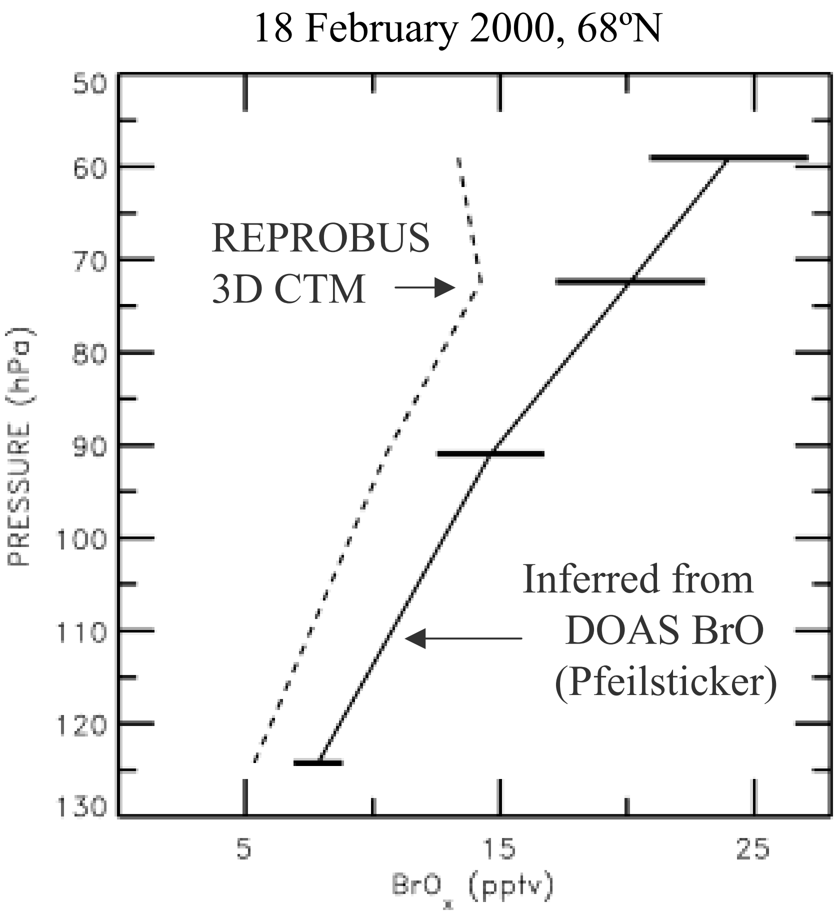
Measured and Modeled Ozone Loss Rates

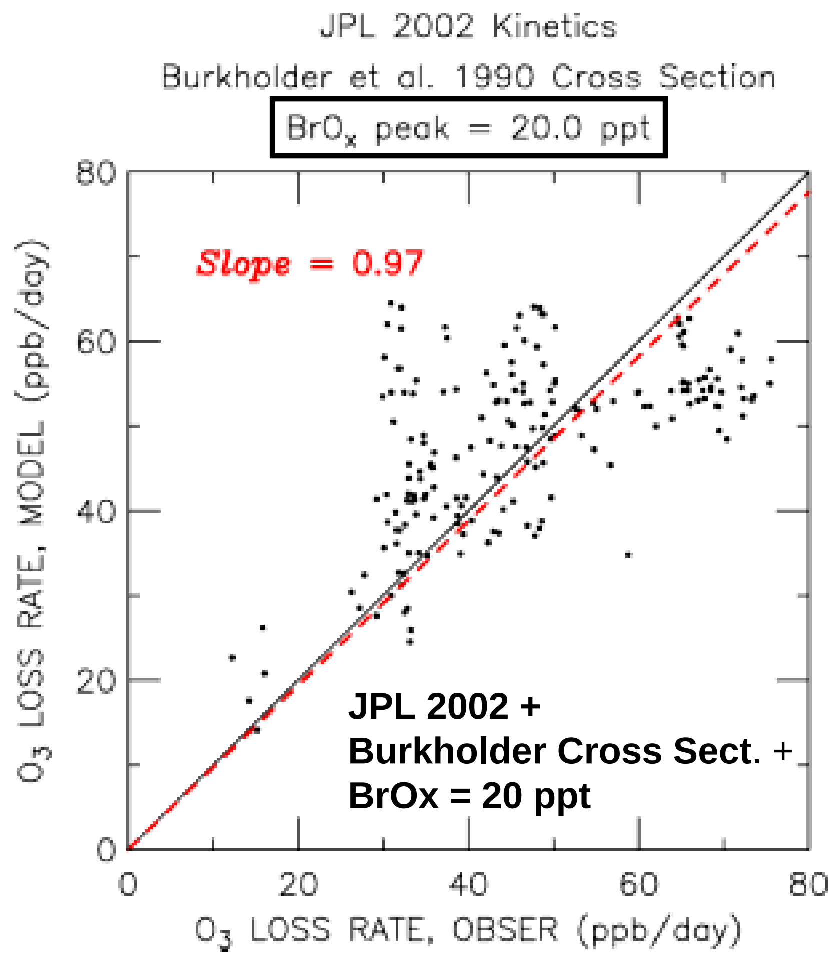




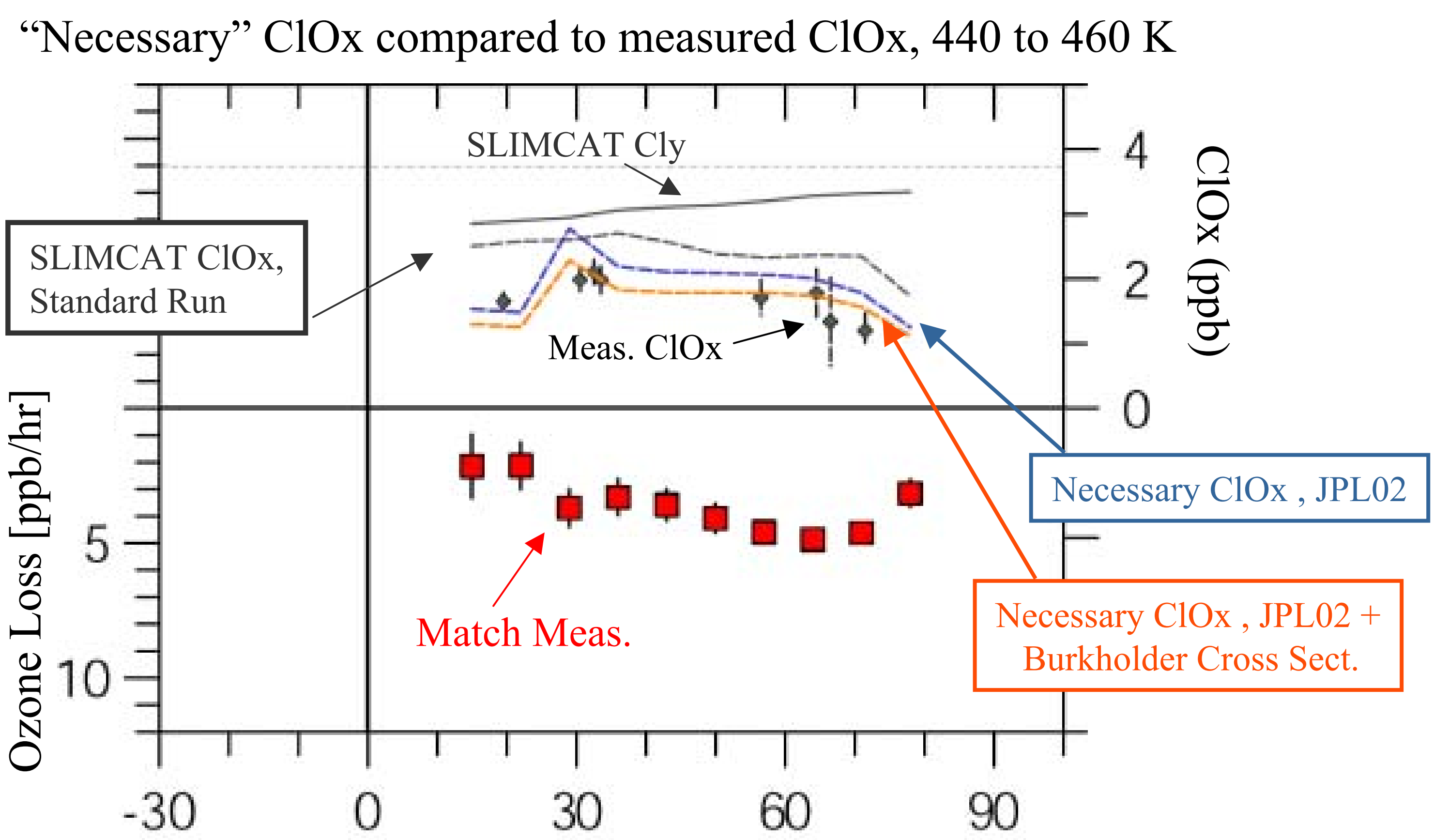
Stimpfle et al.. JGR, 2004

Measured and Modeled Ozone Loss Rates



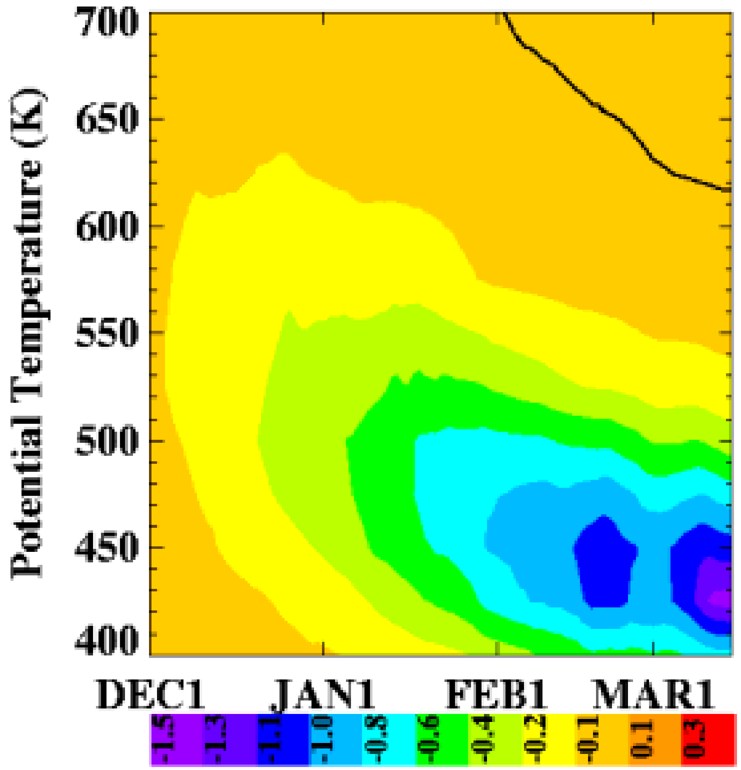


Measured and Modeled Ozone Loss Rates : MATCH

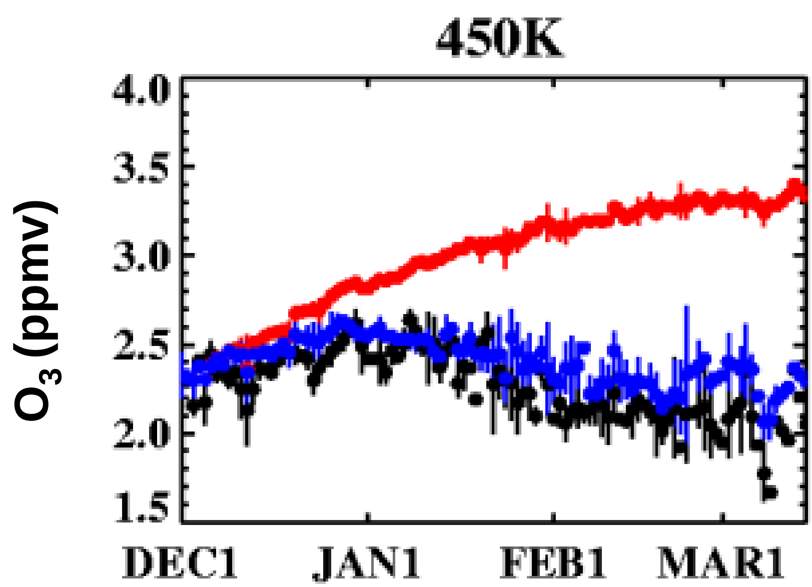


Modeled & Measured Ozone Loss
POAMIII & SLIMCAT
Arctic, 2002-2003

Modeled Ozone Loss:
Active – Pseudo Passive



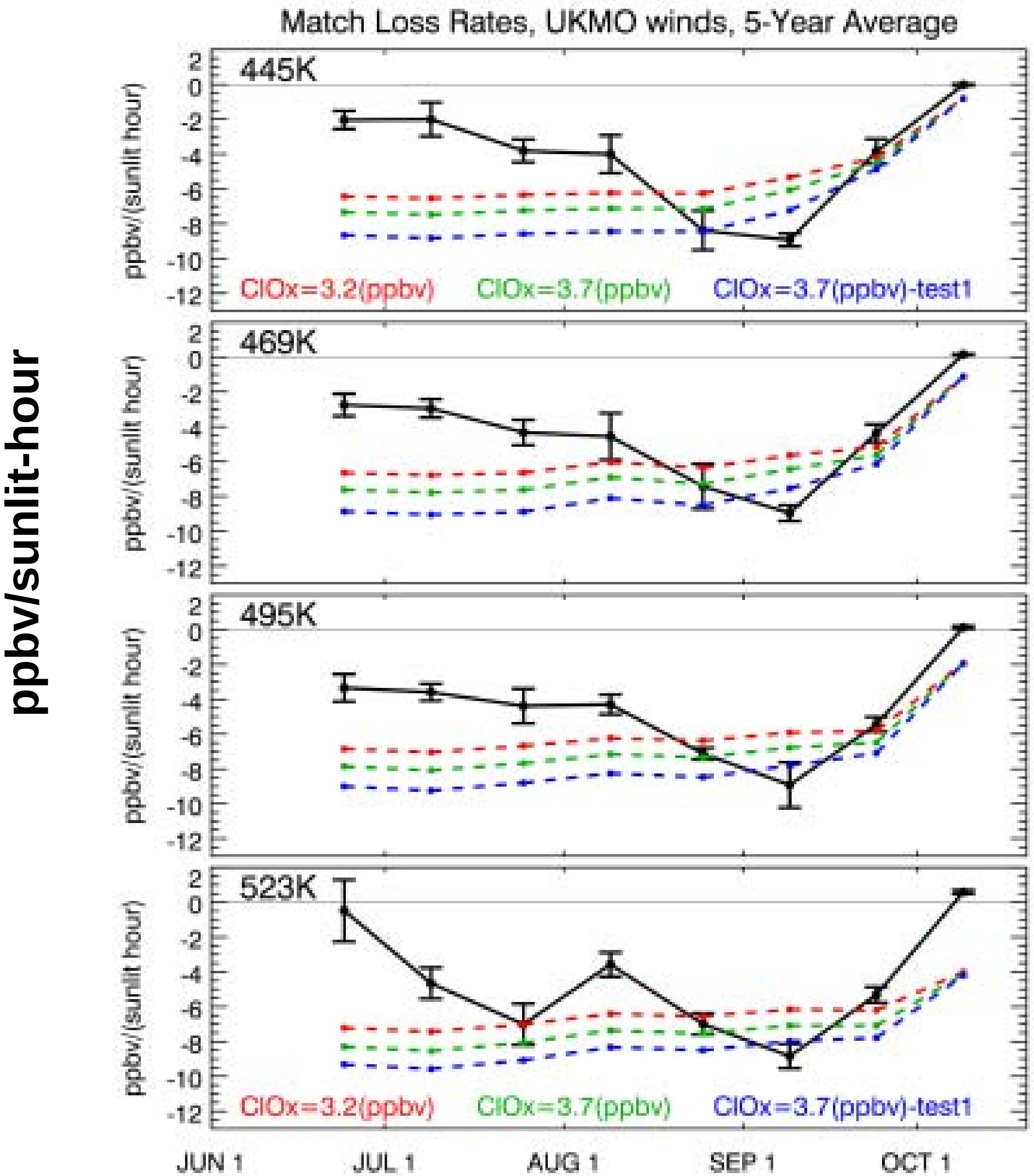
O_3 Difference (ppmv)



POAM III
SLIMCAT Active
SLIMCAT Pseudo Passive

Singleton et al., PP-2

POAM-Match Antarctic Ozone loss rates, 5-Year average (98,99,00,01,03)



Red: photochemical model with **JPL 2002 kinetics**, BrOx=20 ppt, ClOx=3.2 ppb

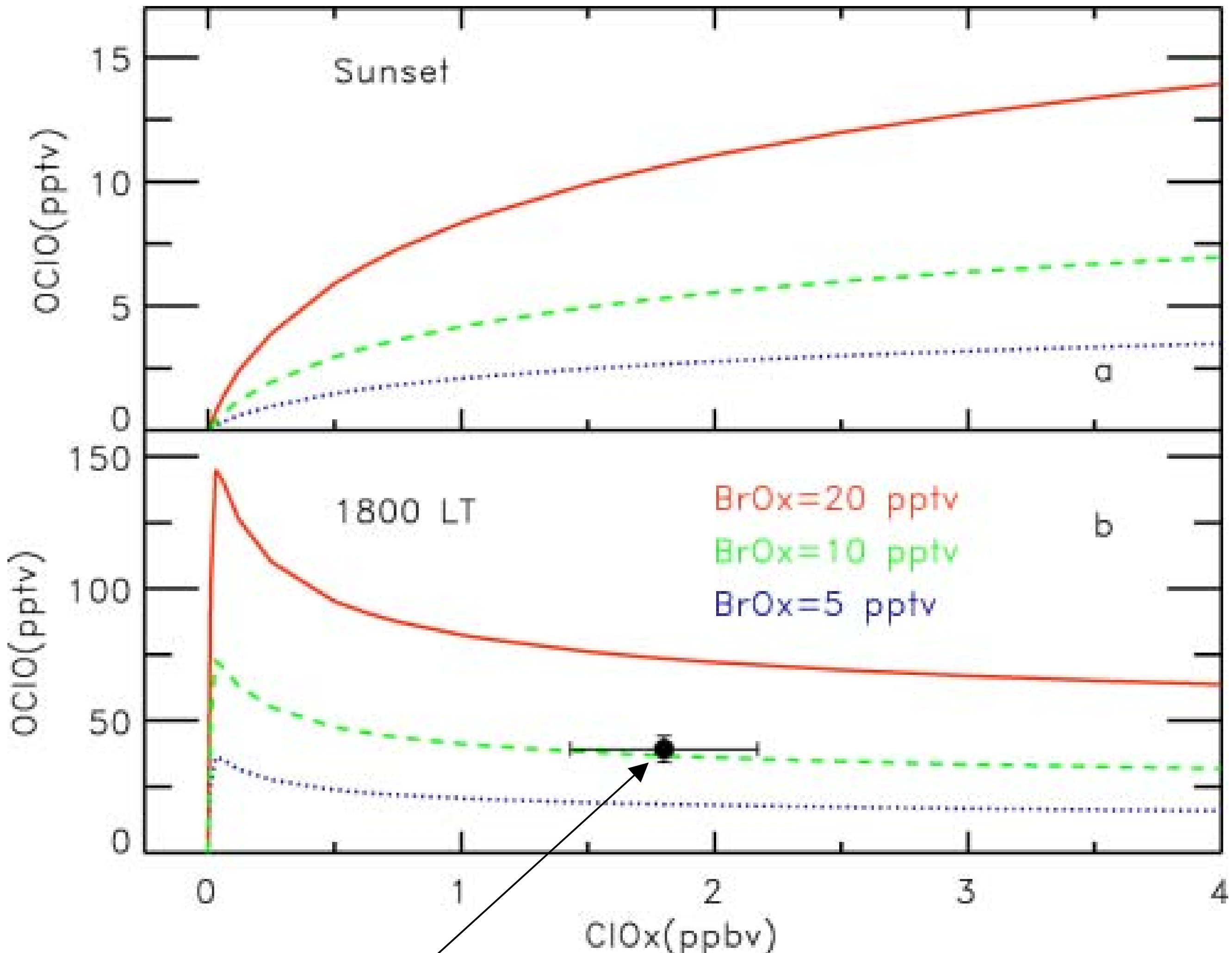
Green: (as above) with **ClOx=3.7 ppb**

Blue: same as green, but with **Burkholder ClOOCl cross sections**

Hoppel et al., PP-1

Nighttime OClO : Indicator of BrO ?!?

JPL 2002 Kinetic Parameters Used In Model



Twilight

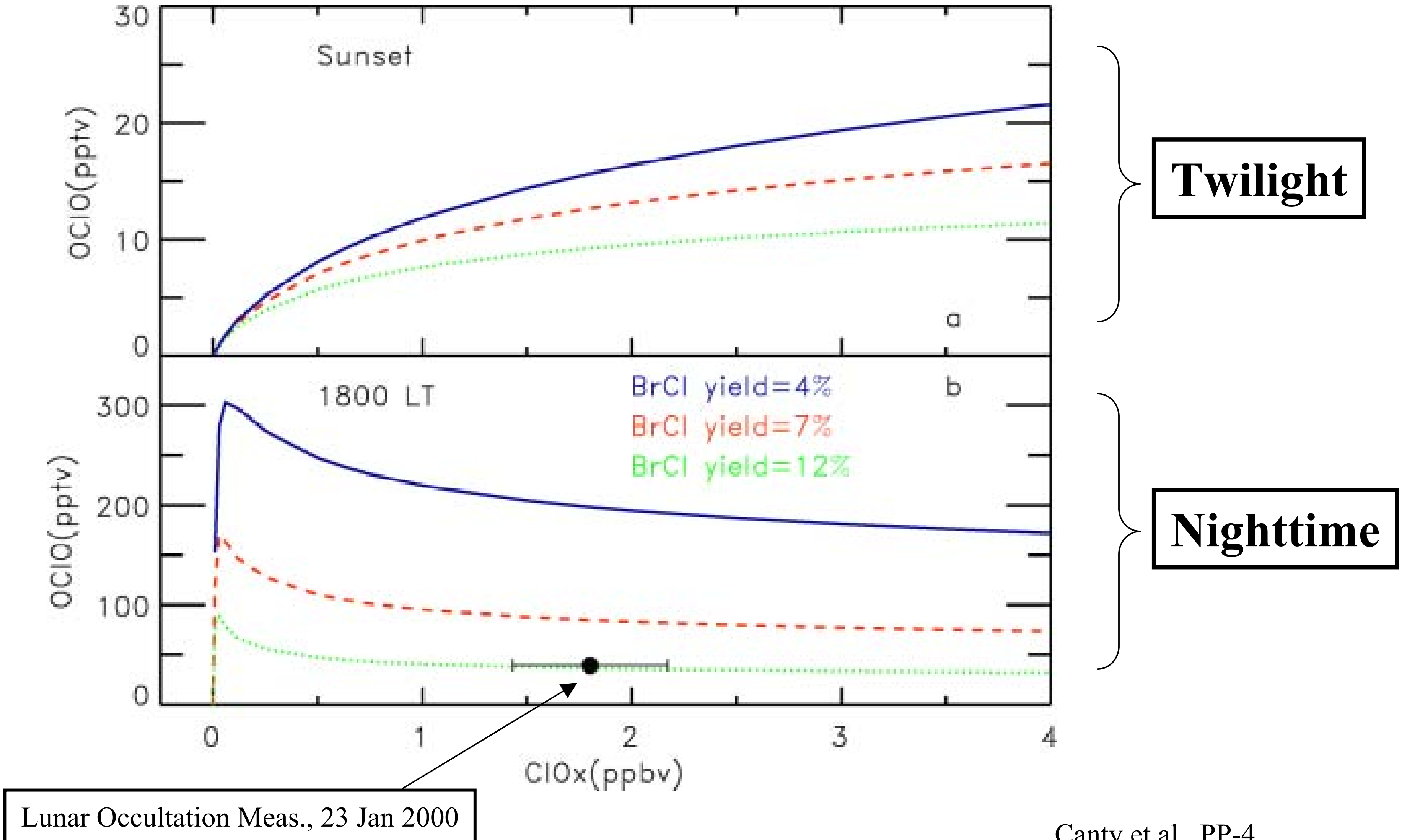
Nighttime

Lunar Occultation Meas., 23 Jan 2000

Canty et al., PP-4

Nighttime OClO : Indicator of BrO ?!?

Model Constrained by BrO_x, inferred from measured BrO, for <u>**various yields of BrO+ClO → BrCl + O_2**</u>



Canty et al., PP-4

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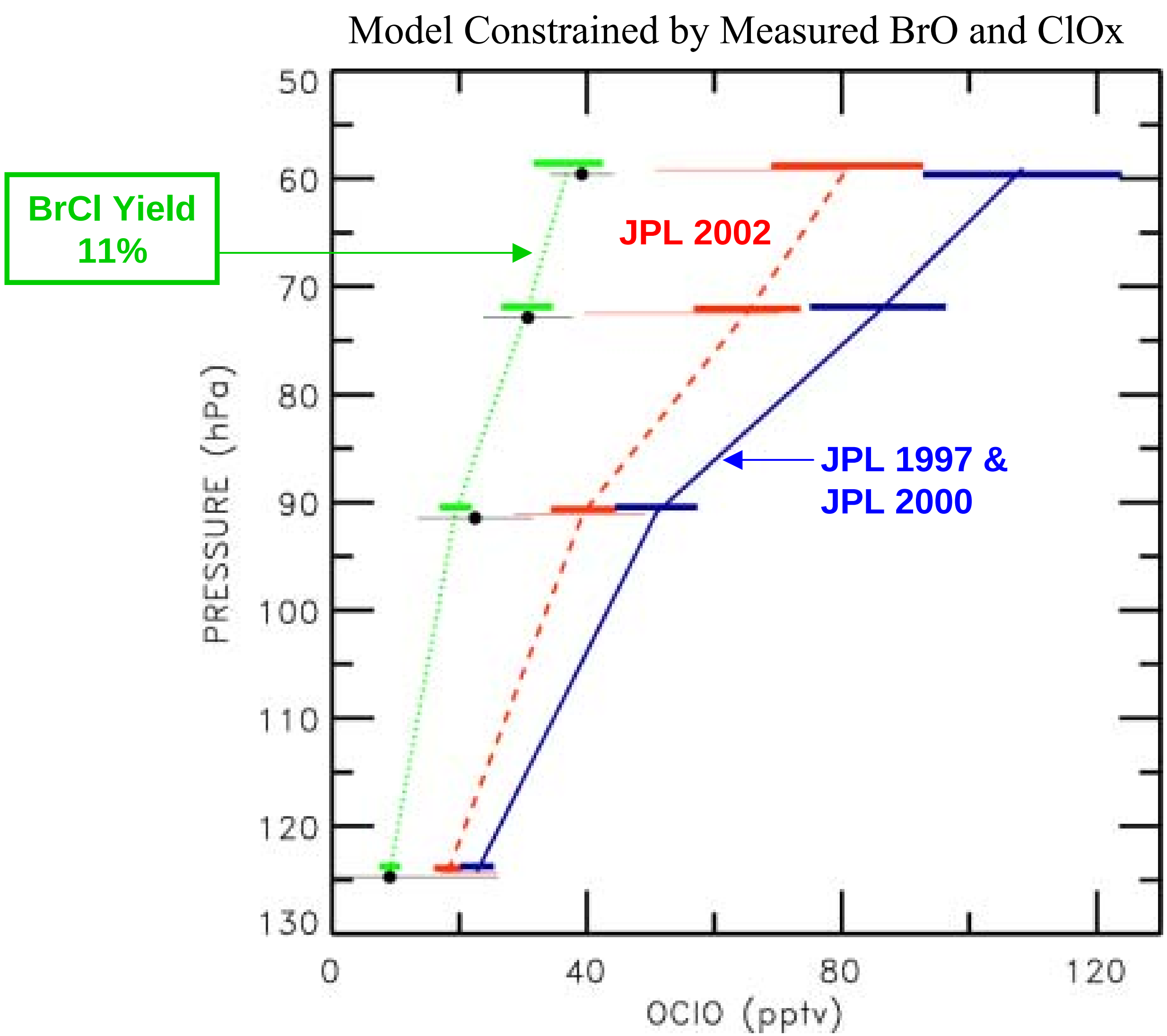
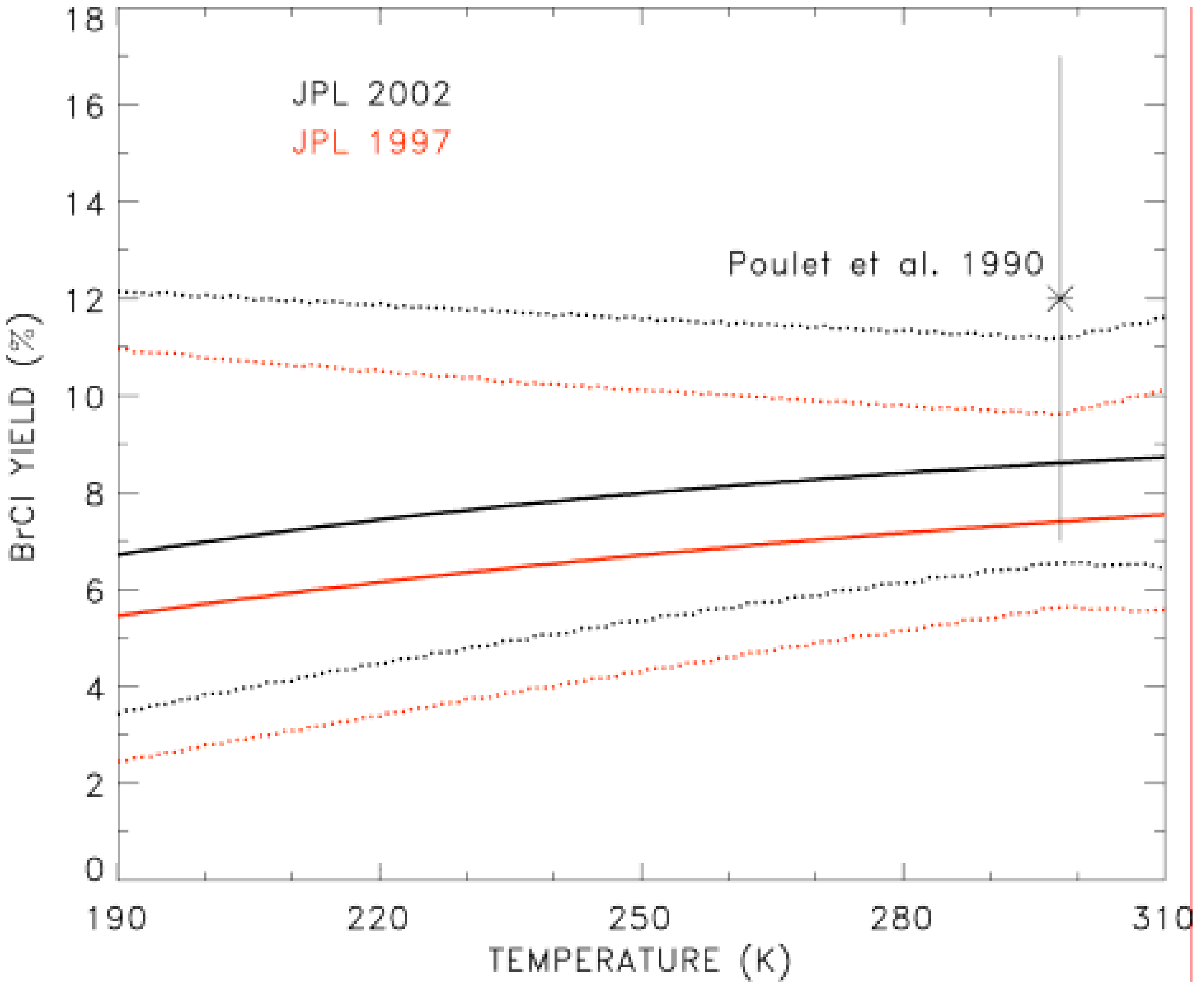


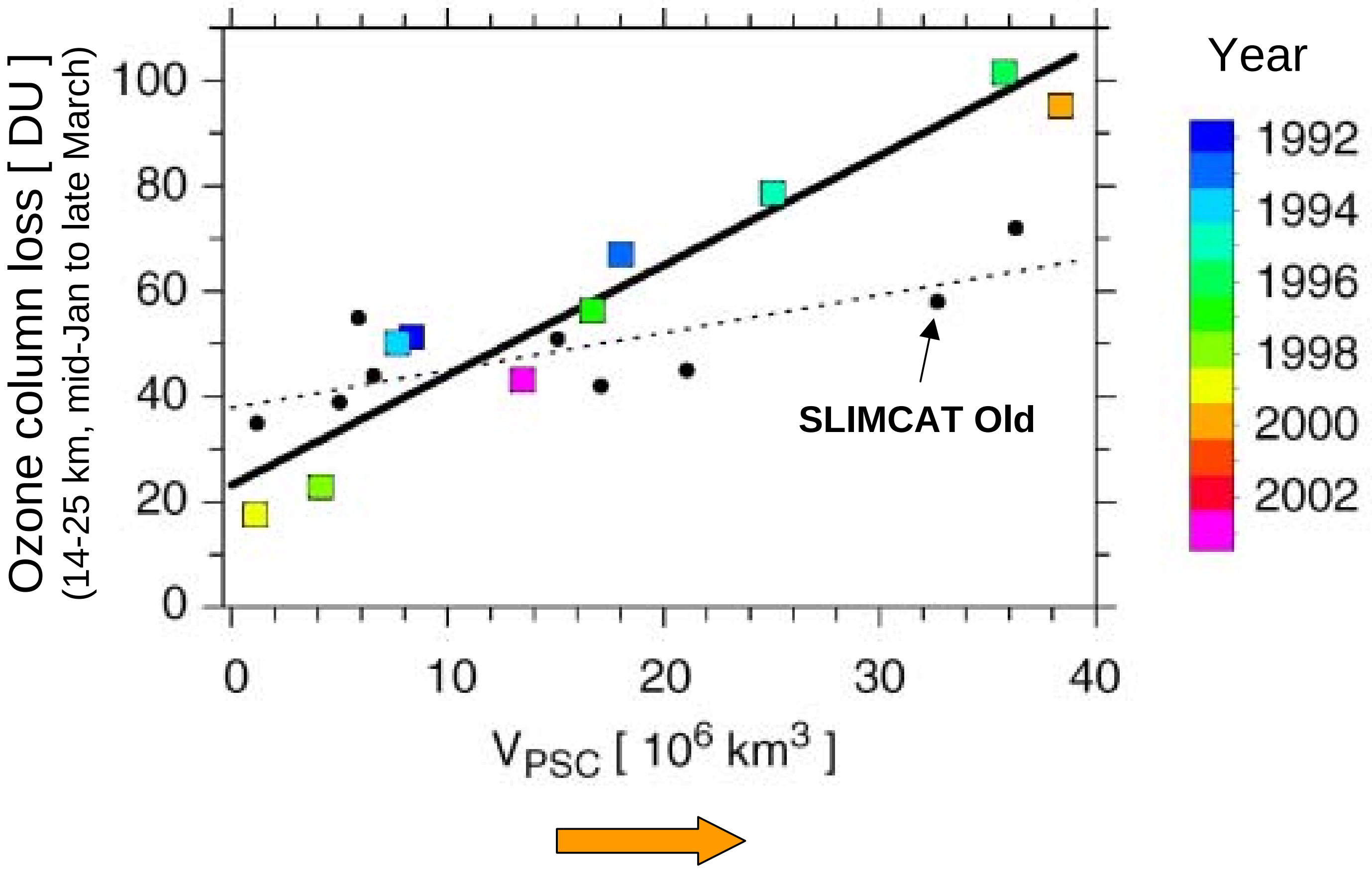
Canty et al., PP-4

BrO + ClO Branching Ratio: Laboratory



Canty et al., PP-4

Comparison with SLIMCAT – Old Version



SLIMCAT "Old" underestimates sensitivity of Arctic ozone loss to climate change by a factor of three

Rex et al., GRL, 2004

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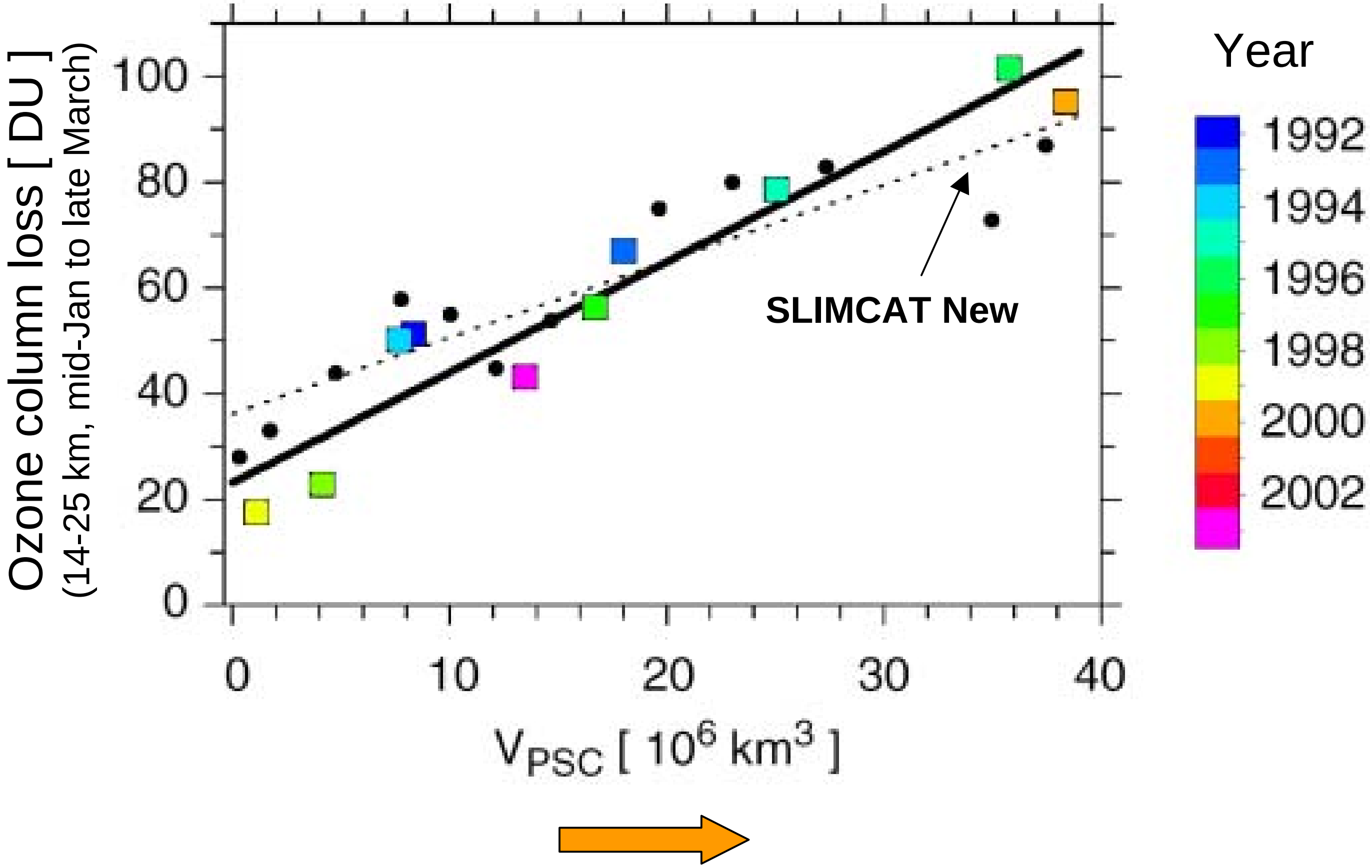


New SLIMCAT version reproduces the slope and scatter of data reasonably well.

New SLIMCAT : JPL 2002 + Burkholder Cross Section + NAT-based Denit. Scheme

Challenges

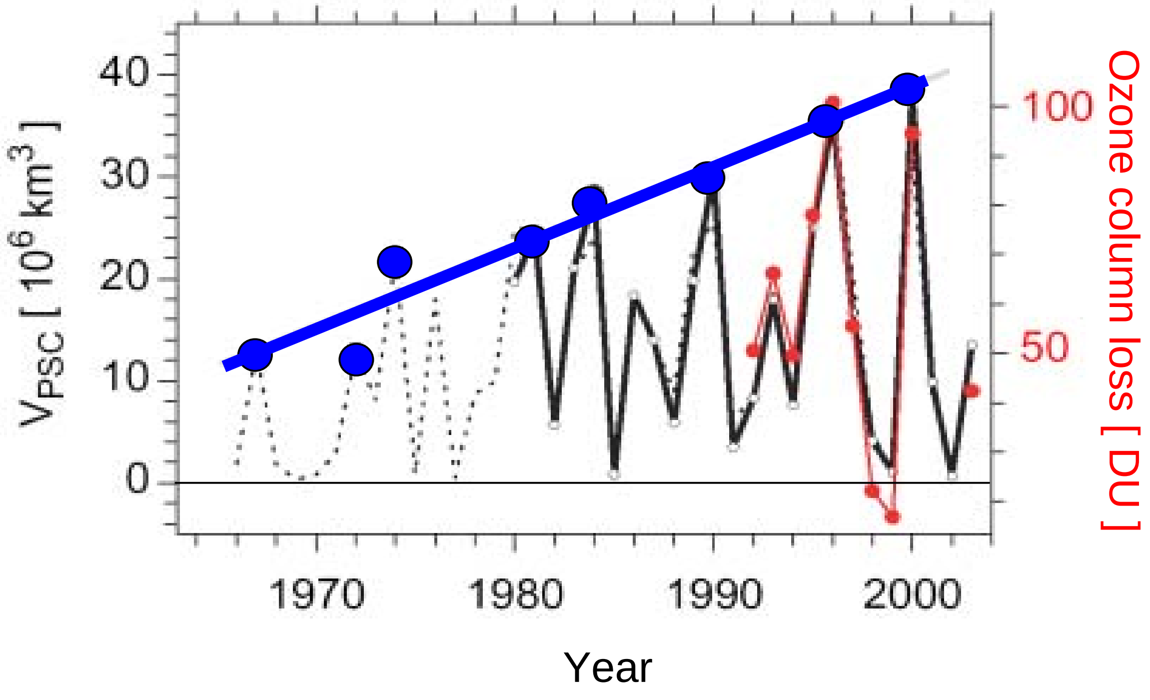
1. Separation of chemistry vs transport using SOSST data
   - column ozone, multiple Arctic winters

2. Measured and modeled ozone loss rates, Antarctic vortex to complement many studies focused on Arctic vortex
   - value added if tied to measured ClO

3. Abundance of BrO in the vortices
   - constraints from nighttime SOSST OClO ?!?

4. Stability of Arctic vortex in a changing climate
   - tests of: dynamical properties (e.g., heat flux vs T)
     transport properties (e.g., tracers) within CCMs
     (Chemistry-Climate Models)

V_{PSC} over the past ~40 years



~ Factor of three increase in max. V_{PSC} over the past four decades

Rex et al., GRL, 2004

PWD (Planetary Wave Drag) and Strength of the Arctic Vortex

$\downarrow$ PWD $\Rightarrow$ $\uparrow$ Arctic Vortex Strength

What is the effect of ↑ GHGs on PWD:

- ↓ PWD due to increased westerly winds in the **subtropics** (Shindell et al. 1998)
- ↓ PWD due to stronger vertical shear of the zonal wind at **high latitudes** (Limpasuvan and Hartmann, 2000)
- ↑ PWD due to weaker vertical shear of the zonal wind at **high latitudes** (Hu and Tung, 2002)
- ↑ PWD due to decreases in the **NAO** (North Atlantic Oscillation) index, driven in part by ↑ **SSTs** from a coupled ocean-atmosphere climate model (Schnadt and Dameris, 2003)

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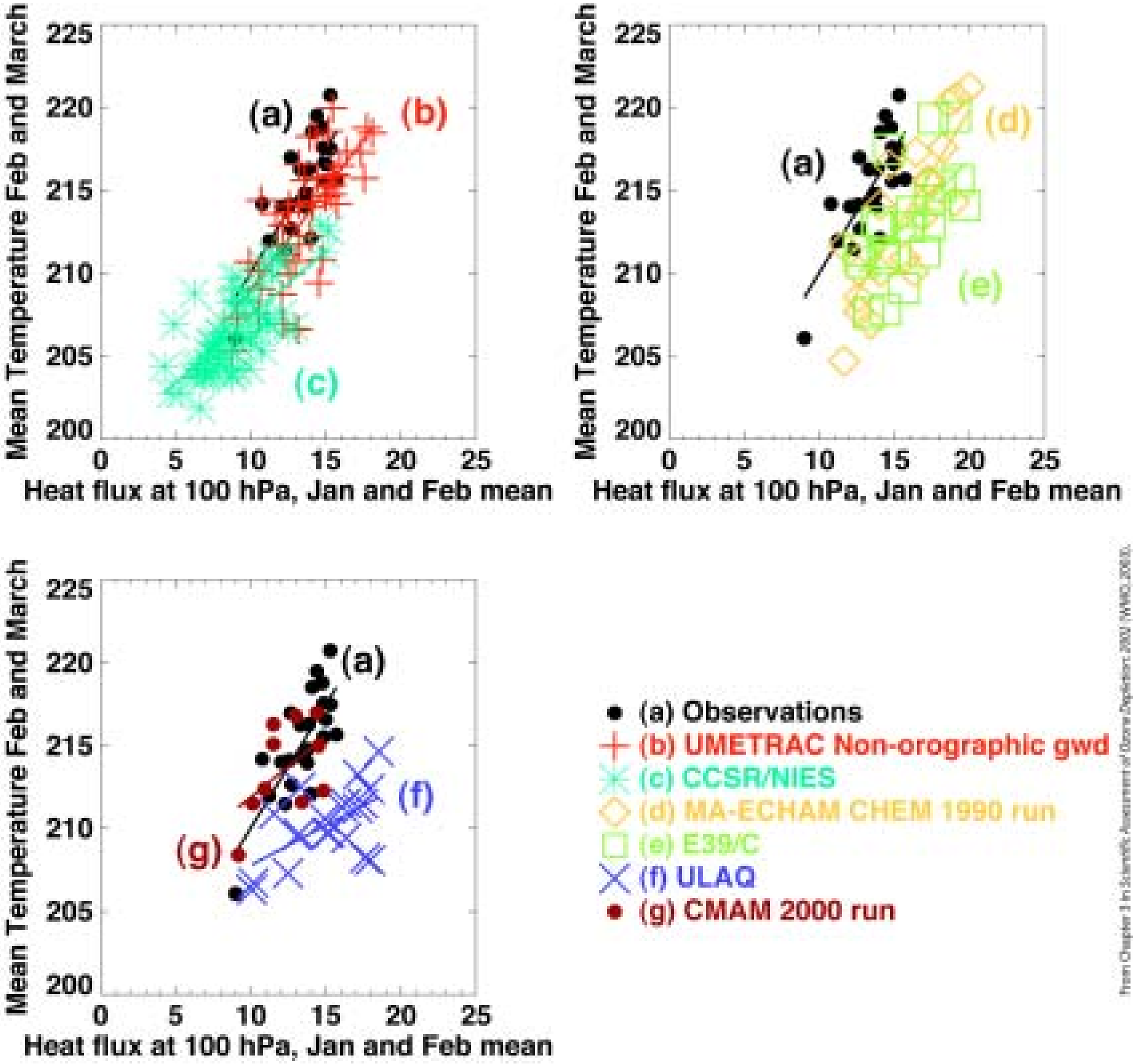
Model evaluation is needed:

- Dynamics: model heat flux (100 mb, Jan-Feb) vs model T (50 mb, Feb-Mar)
  compared to observations: "Newman Plot"
  see Fig 4 of Austin et al., 2003 & Fig 3-43, WMO 2003

- Transport: comparison of modeled and measured tracers, for tracers with
  a variety of lifetimes:
  - SAGE and HALOE O_3 in LS
  - HALOE CH_4
  - Aura N_2O, CFCs, CH_4, O_3
  - Sub-orbital SF_6, CO_2, CH_3Br, etc.

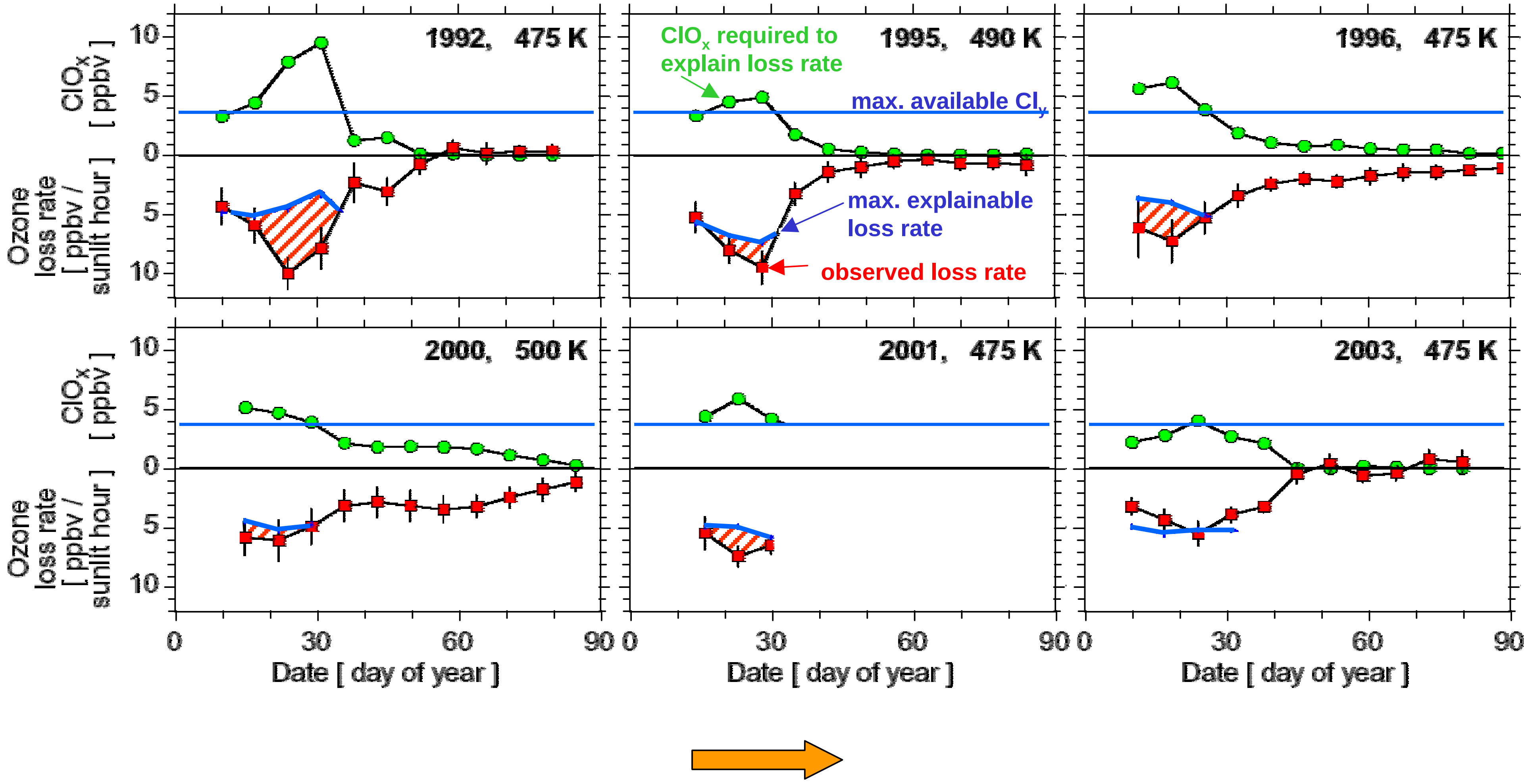
Extra Material To Follow

PWD (Planetary Wave Drag) and Strength of the Arctic Vortex



January ozone loss – model

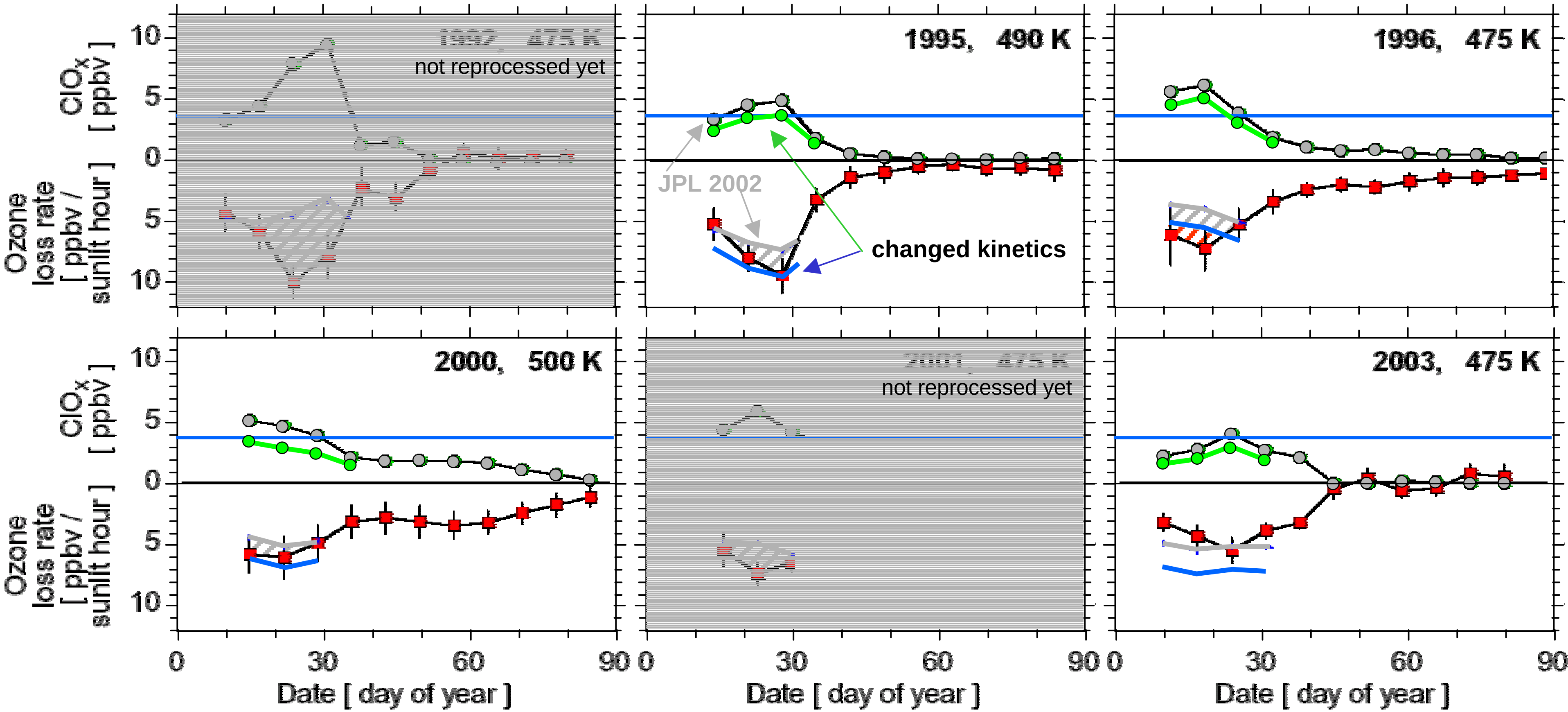
Box model based on ClO_x, BrO_x, O_x chemistry, run along Match trajectories to calculate ClO_x that is required to explain the observed loss rates.



During cold Arctic Januaries ozone loss is consistently faster than can be explained with standard (JPL 2002) reaction kinetics

ClO_x kinetics – results from recent field campaigns

- SOLVE => Cl_2O_2 photolysis faster (Stimpfle et al., JGR 109, 2004)
- EUPLEX => Cl_2O_2 thermal decomposition faster (von Hobe et al., Koch et al., posters 488, 466)
- Here also: ClO + ClO from Bloss et al., BrO_x based on Pfeilsticker et al.



With these changes in reaction kinetics the January ozone loss problem may be largely resolved (see also poster 460, Frieler et al.)

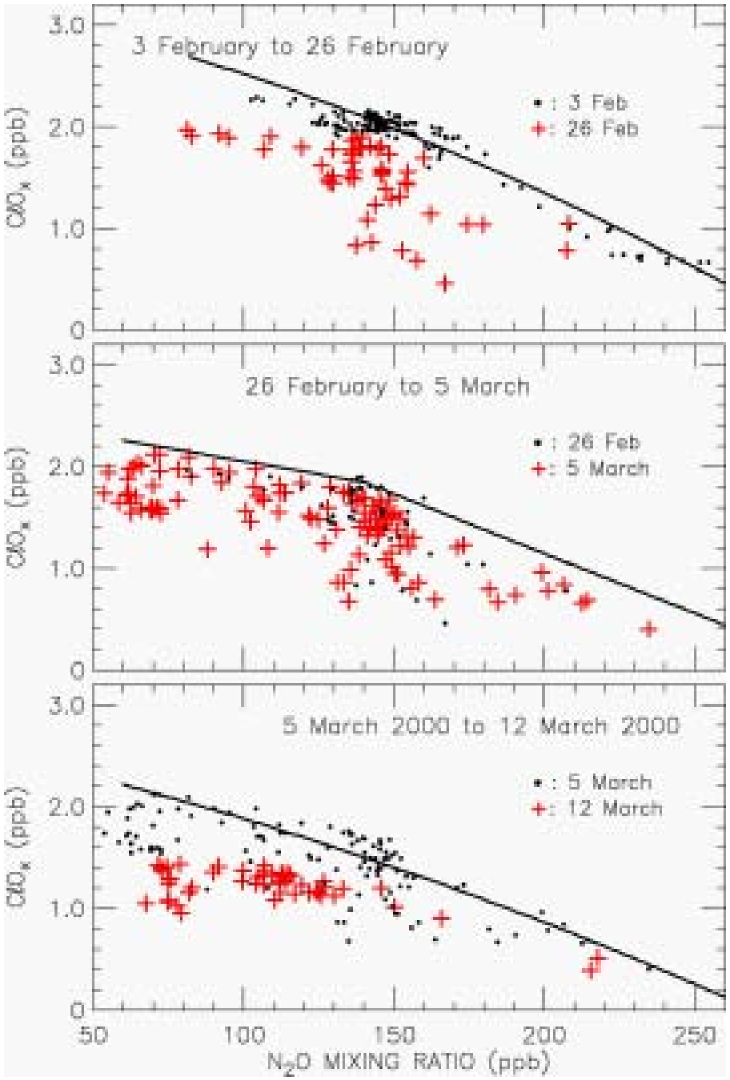
Photochemical Model Description – ClO*x*

- Photochemical model run along back trajectories, originating from the ER-2 flight track, for 10 day periods

- ER-2 observations of ClO*x* and O_3 used to initialize model

- ClO*x* allowed to increase linearly, "backwards in time" to match ER-2 observations obtained at earlier times

ClO*x* ≡ ClO + 2 × ClOOCl



Photochemical Model Description : ClO*x* Partitioning

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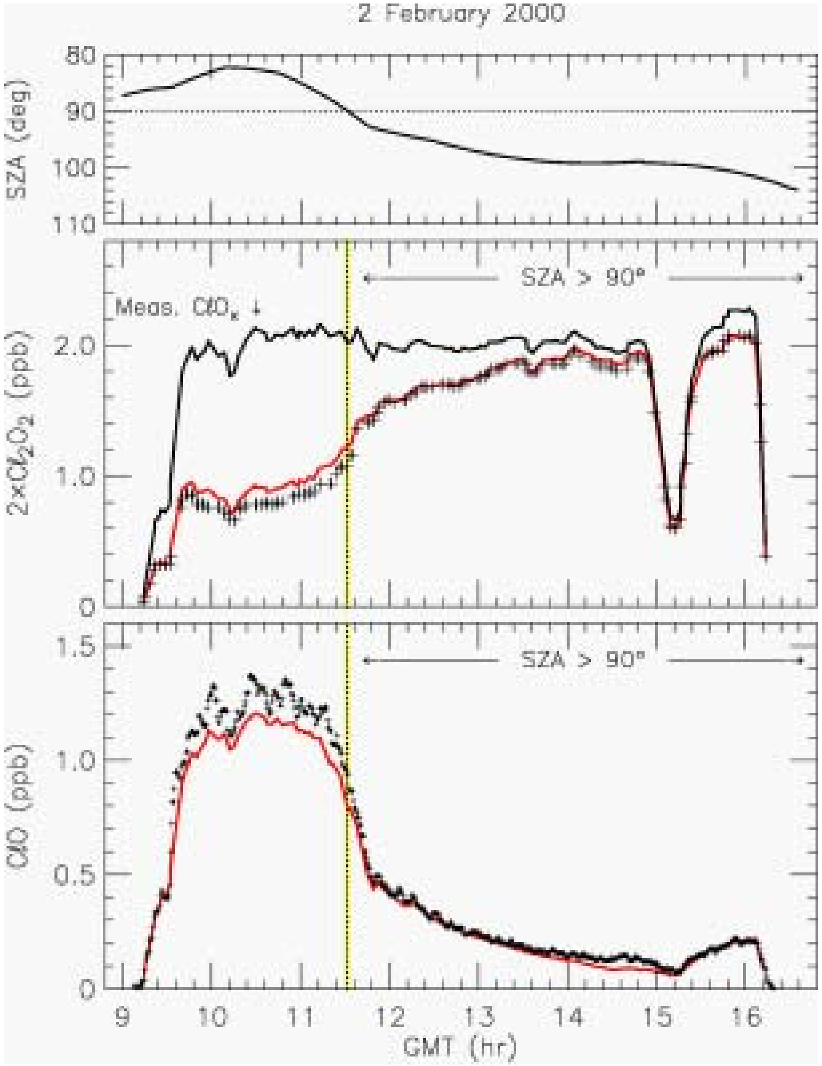
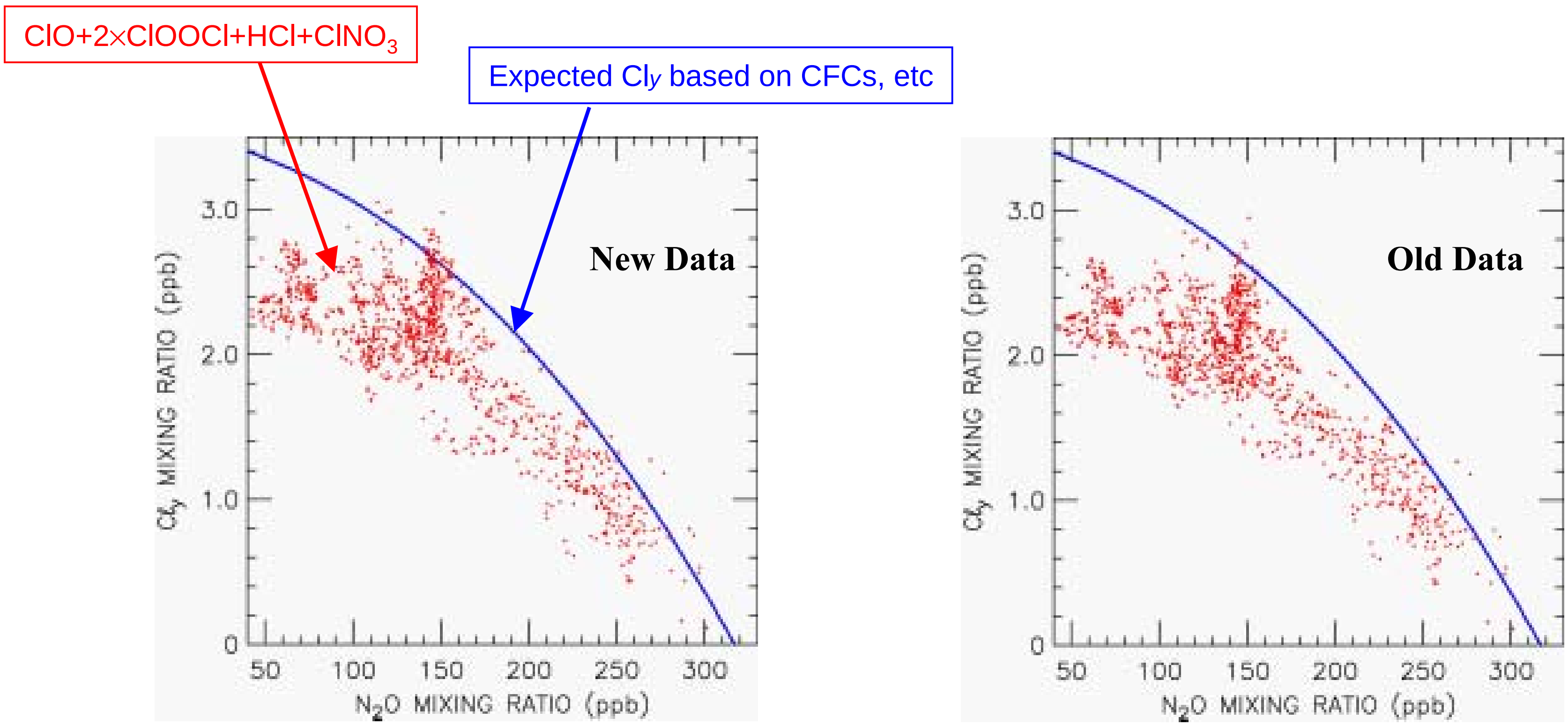
- BrO*x* specified from Pfeilsticker *et al*. DOAS meas. of BrO from Kiruna, winter of 1999/2000

- Model provides reasonably good simulation of the observed partitioning between ClO & ClOOCl along the ER-2 flight track

JPL 2000 Kinetics Used, Unless Otherwise Specified

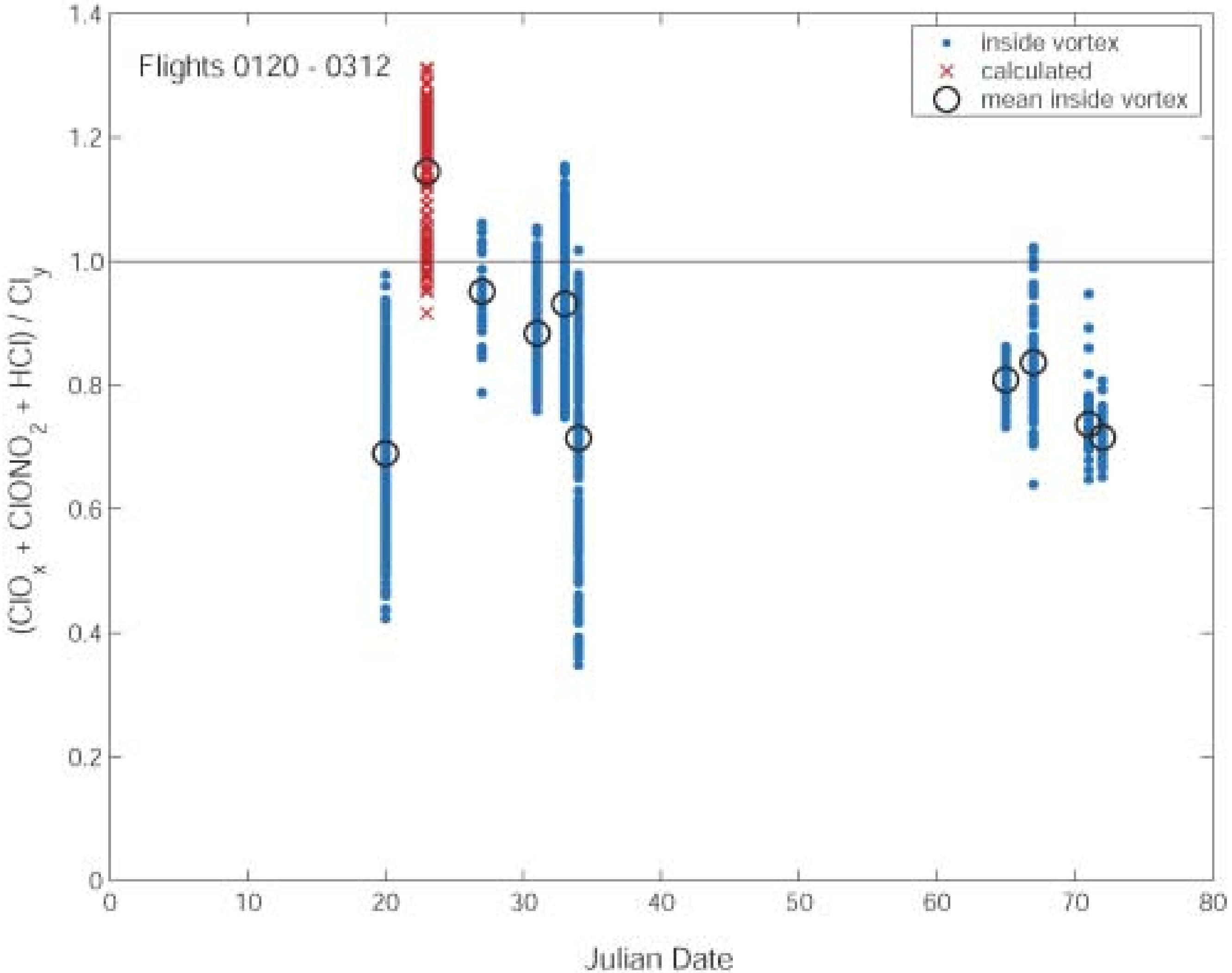


Chlorine Budget



Please see Wilmouth et al. poster for details

Chlorine Budget II



From Wilmouth et al. poster

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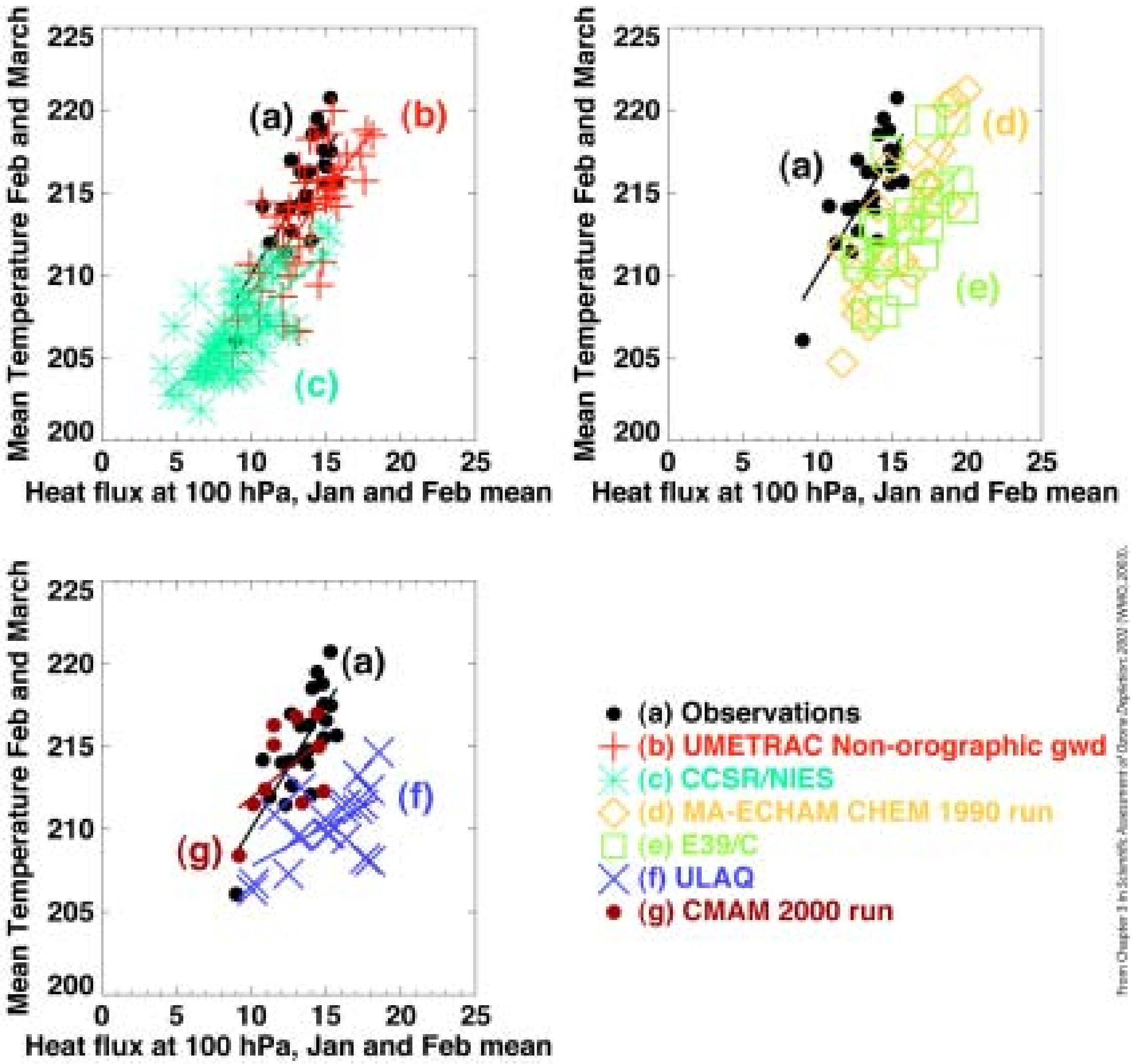

Challenges – Mid-Latitude Ozone

1. Definition of trends in O_3 vs altitude
   - trend quality SOSST O_3 below 20 km

2. Accuracy of tropospheric O_3 retrievals
   - **validation of SOSST tropospheric ozone !!!**

3. Definition of trends in H_2O vs altitude
   - validity of SOSST H_2O for trends?

4. Stratospheric Surface Area Climatology
   - effects of small particles on SSA for background periods

5. Atmospheric Transport
   - tracer fields: CH_4, HF
   - use of O_3, H_2O, SSA as tracers

Challenges – SOSST Future

1. Future measurement needs for stratospheric ozone trends
   - definition of info obtained from various tracers

2. Future measurement needs for tropospheric ozone
   - validity of SOSST O_3
   - measurements to compliment Aura

3. Future measurement needs for water cycle
   - value of H_2O isotopes
   - which tracers needed

4. Other scientific issues: climate change
   - how to improve on SAGE II, HALOE, POAM III